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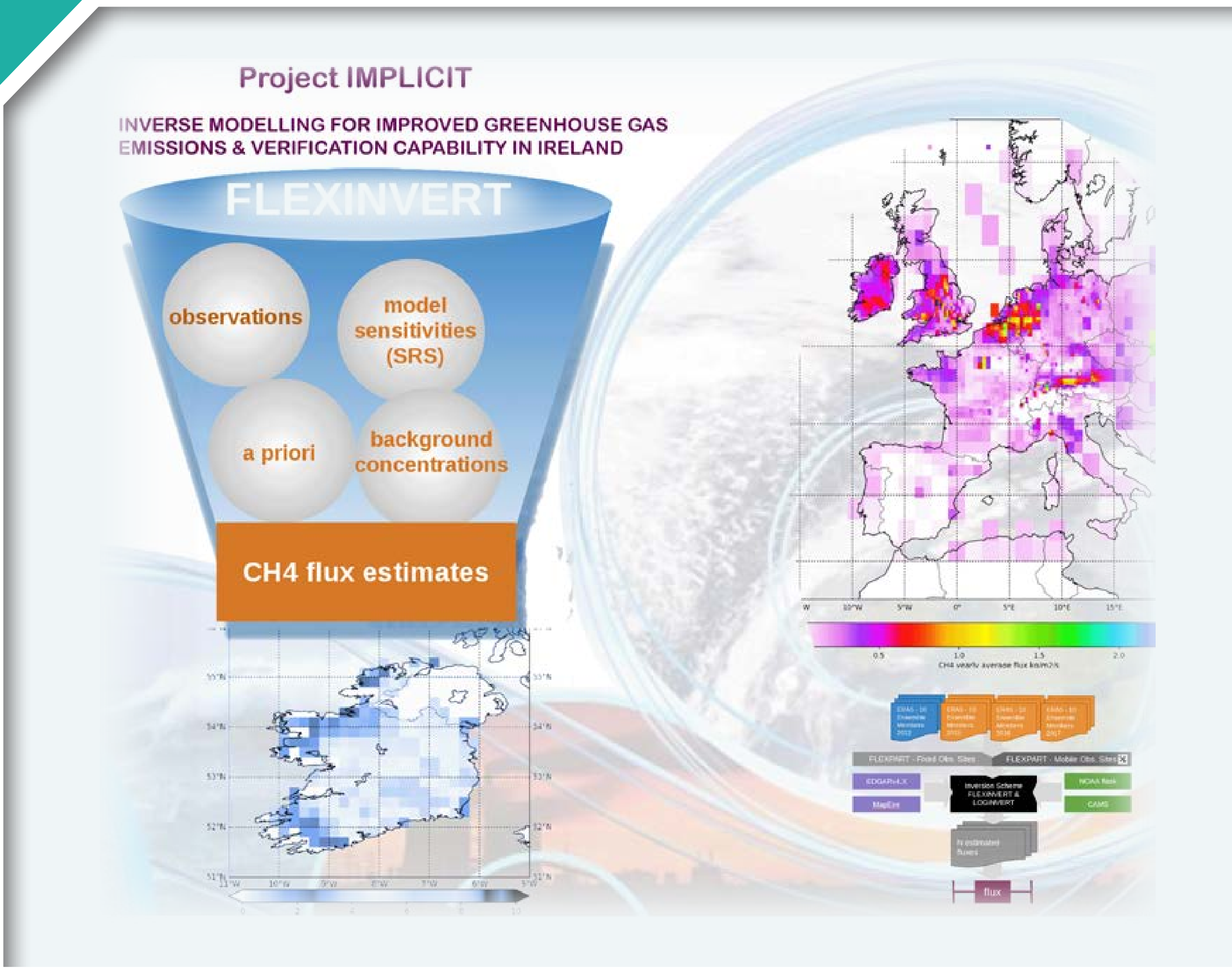
IMPLiCIt: IMProving inversion modeL Capability in Ireland

Authors: Colin O'Dowd, Damien Martin and Dèlia Arnold



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EPA RESEARCH PROGRAMME 2014–2020

IMPLiCIt: IMProving inversion modeL Capability in Ireland

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Prepared for the Environmental Protection Agency

by

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The EPA Research Programme addresses the need for research in Ireland to inform policymakers and other stakeholders on a range of questions in relation to environmental protection. These reports are intended as contributions to the necessary debate on the protection of the environment.

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Executive Summary

The IMPLiCIt (IMProving inversion modeL Capability in Ireland) project has focused on developing capabilities within the Irish state for the inverse modelling of greenhouse gases (GHGs). The IMPLiCIT project involved the design, development and implementation of a system for CH_4 emission verification using the FLEXINVERT (FLEXible INVERTion dispersion model) Bayesian inversion framework (an atmospheric Bayesian inversion framework for determining surface fluxes of trace species using an optimised grid),[1] utilising Ireland's own observational datasets and a priori information, combined with atmospheric transport modelling calculations performed with the well-established FLEXPART (FLEXible PARTicle Lagrangian dispersion) model.

The FLEXINVERT inversion framework has been adapted and prepared for the Irish domain and the first assessments of top-down emissions were conducted for the year 2012. The inversion results show higher (~30%) CH_4 emissions for Ireland compared with two alternative sets of a priori data. These results are likely to be on the margin of the range of the uncertainty of the bottom-up and top-down emissions used here.

Nevertheless, additional work is necessary to characterise and evaluate various different contributors to inversion uncertainty and to implement procedures to reduce and quantify these uncertainties. A number of recommendations are made in order to achieve this:

- Perform multiyear, high temporal resolution analysis to explore the inter-annual and seasonal variation in flux estimates. This would include spike removal on all datasets to preclude any local interference.
- Further investigation of the sensitivity of the inversion system to different meteorological driving data, background definition, a priori data and boundary layer height in the model.
- Constrain the uncertainties related to the atmospheric transport modelling (calculation of the source receptor sensitivities) through ensemble modelling.
- Explore additional measurement sites that would allow, from a sectoral perspective, a better comparison with bottom-up emissions inventories.
- Continue to improve this newly developed capacity to deliver improved flux estimations, which is essential for the delivery of more accurate and verified national emissions inventories for Ireland.

1 Bayesian inference is a method of statistical inference in which Bayes' theorem is used to update the probability for a hypothesis as more evidence or information becomes available. Bayesian inference is an important technique in statistics, and especially in mathematical statistics.

1 Introduction

Atmospheric monitoring of greenhouse gases (GHGs), combined with inverse modelling, can trace back observed atmospheric concentrations of GHGs to their origin, i.e. to the regions where they have been emitted into the atmosphere, and provide top-down estimates of the GHG emissions. This is particularly relevant in the context of the reporting of national GHG emissions to UNFCCC (which are based on bottom-up inventories). (Workshop "Atmospheric monitoring and inverse modelling for verification of national and EU bottom-up GHG inventories", 8–9 March 2007, Ispra, Italy)

1.1 Background

Exposure of the general population to poor air quality can have adverse impacts on public health and the environment. At both national and international levels, countries are encouraged to take the appropriate measures to control emissions and to develop and establish mitigation strategies. This is contingent on the accuracy of emissions inventories. Emissions inventories can be developed through bottom-up, top-down and downscaling approaches (e.g. EEA, 2013; Guevara *et al*., 2014; Kuenen *et al*., 2014), with the bottom-up methodologies being the most common approach. This approach relies on emission factors and activity data, which are updated periodically by different sources. However, these data can change over time and become obsolete and are associated with a number of uncertainties (Hanna *et al*., 2001; Menut and Bessagnet, 2010) that are difficult to address and can ultimately affect the assessments and the associated conclusions derived from them. These uncertainties may include unknowns in the disaggregation of budgets, over-/underestimation of sources, incorrect timing and spatial distribution of the emissions and even the failure to consider undeclared or unknown emissions (Vautard *et al*., 2003; Viana *et al*., 2005; Stohl *et al*., 2013).

Inverse modelling, which combines model output and observational data (ground-based, aircraft-based or satellite), has been developed and used to overcome these limitations and to complement, evaluate and improve existing emissions models (e.g. Bergamaschi *et al*., 2000; Queló *et al*., 2005; Brioude *et al*., 2011; Saide *et al*., 2011). This approach is based on the minimisation of the differences between the modelled concentrations and the observations

Currently, inverse modelling with ground-based measurements has been successfully used to estimate anthropogenic fluxes (e.g. Thompson *et al*., 2015) at global, continental, country and regional scales on a monthly to yearly basis. CH_4 has been one of the target pollutants for such studies as it is an important climate-forcing gas with a large global warming potential. It has a relatively short atmospheric lifetime and is an important contributor to many atmospheric chemistry processes.

1.2 The IMPLiCIt Project

The IMPLiCIt (IMProving inversion modeL Capability in Ireland) project is a research and development service funded by the Irish Environmental Protection Agency (EPA). The project aims to develop a combined measurement and modelling system to verify CH_4 sources over Ireland and regions affecting the Irish domain and to improve Irish national capacities to estimate and verify national CH_4 and other greenhouse gas (GHG) emissions inventories. An inverse modelling system based on the atmospheric transport model FLEXPART (FLEXible PARTicle Lagrangian dispersion model; Stohl *et al*., 2005), together with the methodology and open-source system developed by Thompson and Stohl (2014) and Thompson *et al*. (2015), FLEXINVERT (FLEXible INVERTion dispersion model – http://flexinvert.nilu.no; Figure 1.1) has been implemented and tested for the Irish domain.

FLEXINVERT (and its latest, improved version FLEXINVERT+) is an inverse modelling system based on a Bayesian approach to optimise the estimates of fluxes of atmospheric gases and pollutants by

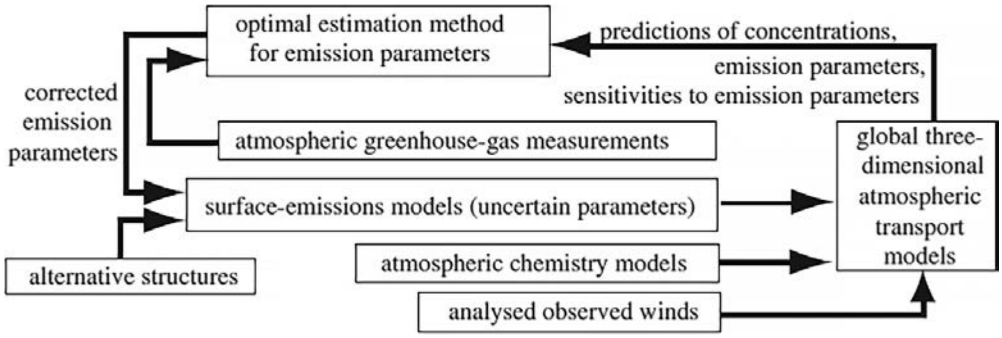

Figure 1.1. Schematic diagram of the FLEXINVERT system, which combines observations, a priori and background information and model sensitivities to provide CH_4 surface flux estimates.

combining the model sensitivities (in the form of source receptor sensitivities or SRSs) obtained by an atmospheric transport model and ground-based measurements. The SRSs describe the relationship between the measurements at the receptor point and receptor time and the emission sources (i.e. fluxes) at the emission location and emission time. In FLEXINVERT, FLEXPART, in a receptor-oriented approach, is used to obtain the SRS fields for a set of selected European measurement stations, including the Irish stations at Mace Head, Malin Head and Carnsore Point, and for 1 year of study (2012).

1.3 Objectives

The primary objective of the project was to improve the inversion modelling capabilities in Ireland applied to GHG emissions, in particular to CH_4, but with a longer term view of potentially expanding this to other pollutants of interest. In brief, the detailed objectives were:

- the implementation, development and optimisation of an inverse modelling system (FLEXINVERT) for CH_4 emissions in the Irish domain;
- the independent verification of emissions and sinks of CH_4 in Ireland based on data from key boundary sites to produce estimates for 1 or 2 years;
- the provisional assessment of the relative contributions from individual sources using modelling and observational data analysis techniques;
- the expansion of expertise in Ireland on inverse modelling of emission estimates; and, finally,
- the establishment of engagement with the community to ensure that the best practices are implemented and provide the starting point for future project collaborations on modelling and assessment of GHG emissions in Europe.

2 Inversion Modelling Framework

2.1 FLEXINVERT

FLEXINVERT is a Bayesian inversion framework developed to optimise surface-to-atmosphere fluxes of an atmospheric species that has either no chemistry or a simple linearised chemistry, based on atmospheric measurements at mobile or fixed stations. FLEXINVERT was initially developed principally for use with GHGs, such as CH_4 or halocarbon species. The code has been and is still being developed by Rona Thompson at the Norsk institutt for luftforskning (Norwegian Institute for Air Research – NILU), Norway. Its continuous evolution is based on the principle of having FLEXINVERT as an adaptable inversion framework that can accommodate different type of problems, species, and temporal and spatial scales. For example, the latest version of FLEXINVERT is able to solve CO_2 fluxes, including resolving the very important diurnal cycle of this species.

This framework is based on the SRSs (also called model sensitivities or transfer coefficient matrices). The SRSs provide an indication of the influence that a given emission source has had on the observed concentration. The SRSs describe the relationship between changes in the mixing ratios at the receptor point and the changes in the fluxes.

FLEXINVERT is based on the Bayes' theorem, with the most probable solution for *x* being the one that minimises the difference between observed and modelled mixing ratios, constrained by the a priori knowledge of the input parameters, including statistical properties (Tarantola, 2005). In FLEXINVERT it is assumed that uncertainties have a Gaussian probability density function that is included in the cost function to be minimised. The cost function can be defined as follows:

$$J(x) = 1/2(x - xb)^{TB-1}(x - xb) + 1/2(Hx - y^{obs})^{T}R^{-1}(Hx - y^{obs}) \quad (2.1)$$

where *x* is a state vector of dimension N × 1, *xb* is an a priori state vector of fluxes and background mixing ratio scalars, *B* (N × N) is the a priori flux error covariance matrix, *H* is the complete atmospheric transport operator of dimension M × N and y^{obs} is a vector of the observed mixing ratios.

A Gaussian approximation allows FLEXINVERT to find an analytical solution to the minimisation problem, that is, to find the *x* for which the first-order derivative of the cost function is zero.

FLEXINVERT has previously been used to estimate fluxes of CH_4, SF_6, HFC-125 and HFC-134a in Europe as part of the European Union-funded InGOS (Integrated non-CO_2 Greenhouse gas Observing System) project; fluxes of CH_4 in East Asia as part of the Norwegian Research Council (NFR)-funded SOGG-EA (Sources of Greenhouse Gases in East Asia) project; and fluxes of CH_4 and black carbon in the Arctic as part of the NFR-funded SLICFONIA (Emissions of Short-lived Climate Forcers near and in the Arctic) project.

FLEXINVERT is not meant to be used as a black box. Evaluation of the species of study, including long-lived species (and therefore with relevant background concentrations) that are chemically reactive, subject to deposition and other parameters, should be taken into consideration when setting up the system and understanding whether FLEXINVERT, or its successor, FLEXINVERT+, should be the system of choice. The a priori information and associated uncertainties, spatial and temporal correlation lengths, and observations will influence the FLEXINVERT output. In summary, FLEXINVERT is aimed at research studies in which a reasonable amount of time and resources can be invested in understanding the system behaviour with respect to the input data and set-up.

FLEXINVERT is coded in Fortran 90 and tested with the GFortran compiler under a Linux operating system. To run FLEXINVERT, the LAPACK and NetCDF libraries for Fortran must be installed. The current version of FLEXINVERT can be run with output from FLEXPART v9.2 (for other versions some modifications may be needed). The recent FLEXINVERT+ requires the usage of FLEXPART v10.

For the IMPLiCIt project a number of modifications were made to the code:

- adaptation of the reading routines to properly read the data provided by the World Data Centre for Greenhouse Gases (WDCGG), in addition to data from the Irish sites;

- adaptation of the reading routines for the background to be able to read in the National Oceanic and Atmospheric Administration (NOAA) flask data provided by Rona Thompson (NILU, personal communication, 15 November 2017), in addition to formatting for future applications with the Copernicus Atmosphere Monitoring Service.

FLEXINVERT has a wide range of options that can be modified by the user to better suit the requirements of the study. These include the selection of a time window for mountain and non-mountain sites, an error for the observations (alternatively, an error per site and observation could be provided with minor modifications), errors for the background data, as well as a total error in the estimated total flux in the inversion, and the spatial and temporal correlation lengths (in km and days respectively), and whether or not to use aggregated grids for those areas where the inversion is not performed at the maximum resolution.

2.2 Observational Data

The station data used to perform the inversions were obtained directly from the WDCGG (see https://gaw.kishou.go.jp/) for all of the stations except the three Irish stations of Mace Head, Malin Head and Carnsore Point; the data for the Irish stations were provided by the National University of Ireland Galway (NUIG). The observational error of individual measurements can be included if the data are available. In this study, fixed error values of 2 or 5 ppb (two tests were carried out) were used. The observation error covariance matrix includes not only the measurement errors but also the much larger transport errors and therefore the influence of measurement uncertainty is expected to be minimal. Table 2.1 and Figure 2.1 present an overview of the available data and the range of concentrations.

For testing, the observational sites were divided into four groups (Figure 2.2):

1. all_sites – all sites ($n = 16$);
2. subset_sites – subset of 12 sites;
3. ie_uk_sites – this subset includes all of the UK and Irish sites;
4. ie_uk_withoutcarnsore_sites – this subset is the same as that above but without Carnsore Point to try to identify how a single station can affect the estimates.

Table 2.1. Data availability according to site

Station full name[a]	Abbreviation	Month[b]											
		1	2	3	4	5	6	7	8	9	10	11	12
Hohenpeissenberg (Germany) – event	HPB	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y
Jungfraujoch (Germany)	JFJ	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y
Monte Cimone (Italy) – event	CMN	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y
Neuglobsow (Germany)	NGL	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y
Ochsenkopf (Germany) – event	OXK	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y
Plateau Rosa (Italy)	PRS	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y
Puy de Dôme (France)	PUY	X	X	X	Y	Y	Y	Y	Y	Y	Y	Y	Y
Ridge Hill (UK)	RGL	X	X	Y	Y	Y	Y	Y	Y	Y	Y	Y	X
Schauinsland (Germany)	SSL	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y
Sonnblick (Austria)	SNB	X	X	X	X	Y	Y	Y	Y	Y	Y	Y	Y
Tacolneston (UK)	TAC	X	X	X	X	X	X	Y	Y	Y	Y	Y	X
Zugspitze (Germany)	ZSF	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y
Mace Head (Ireland)	MHD	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y
Carnsore Point (Ireland)	CRN	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y
Malin Head (Ireland)	MAH	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y

[a]Those marked as "event" are those with sparse data.

[b]Red indicates lack of available data.

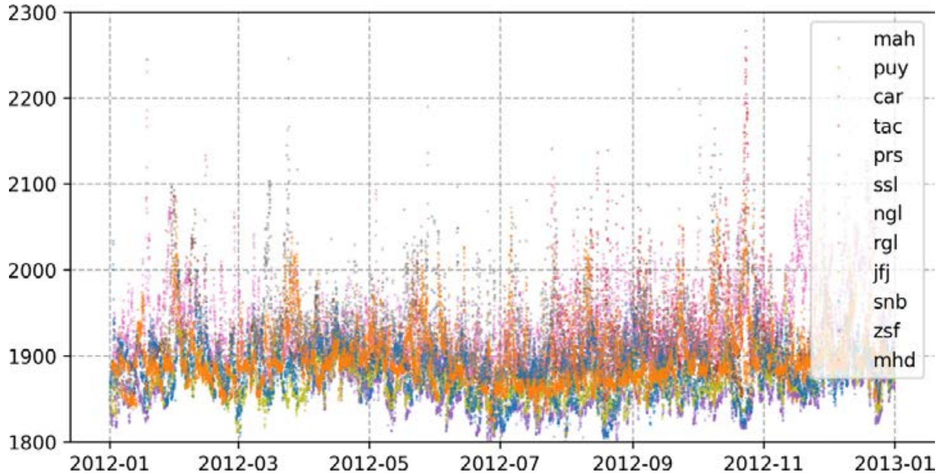

Figure 2.1. Time series of the observations used in the inversions. The plot is cluttered and should be used not to identify the independent observations but to understand the overall variations and ranges.

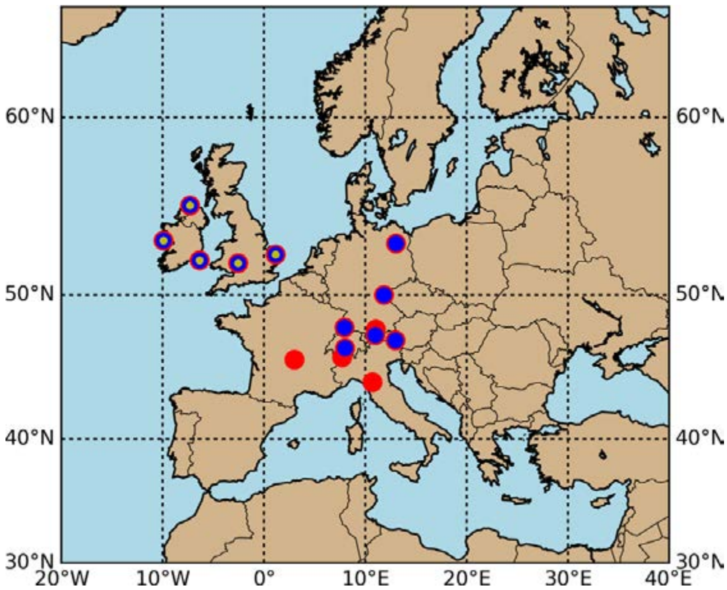

Figure 2.2. Map showing the stations used in the inversion according to subset. All sites – *n* = 16; subset_sites – *n* = 12 (all sites with red dots subtracted); ie_uk_sites – this subset includes all of the UK and Irish sites.

2.3 A Priori and Background Information

The inversion modelling system implemented in the IMPLiCIt project depends on the balance of observations, a priori and background data and their corresponding errors. In the IMPLiCIT project, two a priori datasets were used:

1. The a priori flux used in Thompson and Stohl (2014), kindly made accessible by Rona Thompson. This flux is based on public data: the Emissions Database for Global Atmospheric Research (EDGAR) for anthropogenic emissions and various sources for natural emission sources (Thompson and Stohl, 2014). All fluxes are at a 1 × 1 degree horizontal resolution, on a monthly basis, and add up to a total of 610 Tg/year. These data will be called EDGAR_1.0×1.0_NAT hereafter.
2. A second EDGAR inventory, EDGARv4.3.2, at a resolution of 0.1 × 0.1 degree, was also used. However, this high-resolution inventory does not include natural sources, which can be significant for CH_4 emissions. In addition, this inventory is based on annual averages, which precludes the monthly resolved information provided by

Thompson's inventory. It should be noted that the inversion, provided that enough observational data are available, should constrain the fluxes and correct them (increase or reduce) where necessary, accounting for sources not originally appearing in the a priori dataset. The EDGAR inventory includes anthropogenic emissions from agriculture, industrial processes, residential and transport emissions, waste and oil, coal and gas emissions. In the high-resolution EDGARv4.3.2 inventory, the emissions are per sector and country. For the energy-related sectors, the activity data are based mainly on the energy balance statistics of the International Energy Agency (IEA, 2014), whereas the activity data for the agricultural sectors originate mainly from the Food and Agriculture Organization of the United Nations (see http://faostat3.fao.org/faostat-gateway/go/to/download/Q/*/E). These a priori data are labelled as EDGAR_0.1×0.1_ANT.

Looking at the original emissions inventories (Figures 2.3 and 2.4, which show July as an example), it can be seen that, despite the resolution difference, the fluxes are of a similar magnitude. In both inventories the fluxes are spatially compatible with the Irish land use, as obtained from the Corine land cover classes (CLC2012) dataset (Figures 2.5 and 2.6).

Errors in the a priori flux estimates are correlated in space and time owing to corresponding correlations in the biogeochemistry model, upscaling model or anthropogenic emission inventory that was used to produce these estimates. Most often, little is known about the true temporal and spatial error correlation patterns. Here, we define the spatial error correlation for the fluxes as an exponential decay over distance. The full temporal and spatial correlation matrix is estimated by the Kronecker product of the two temporal and spatial error correlation matrices.

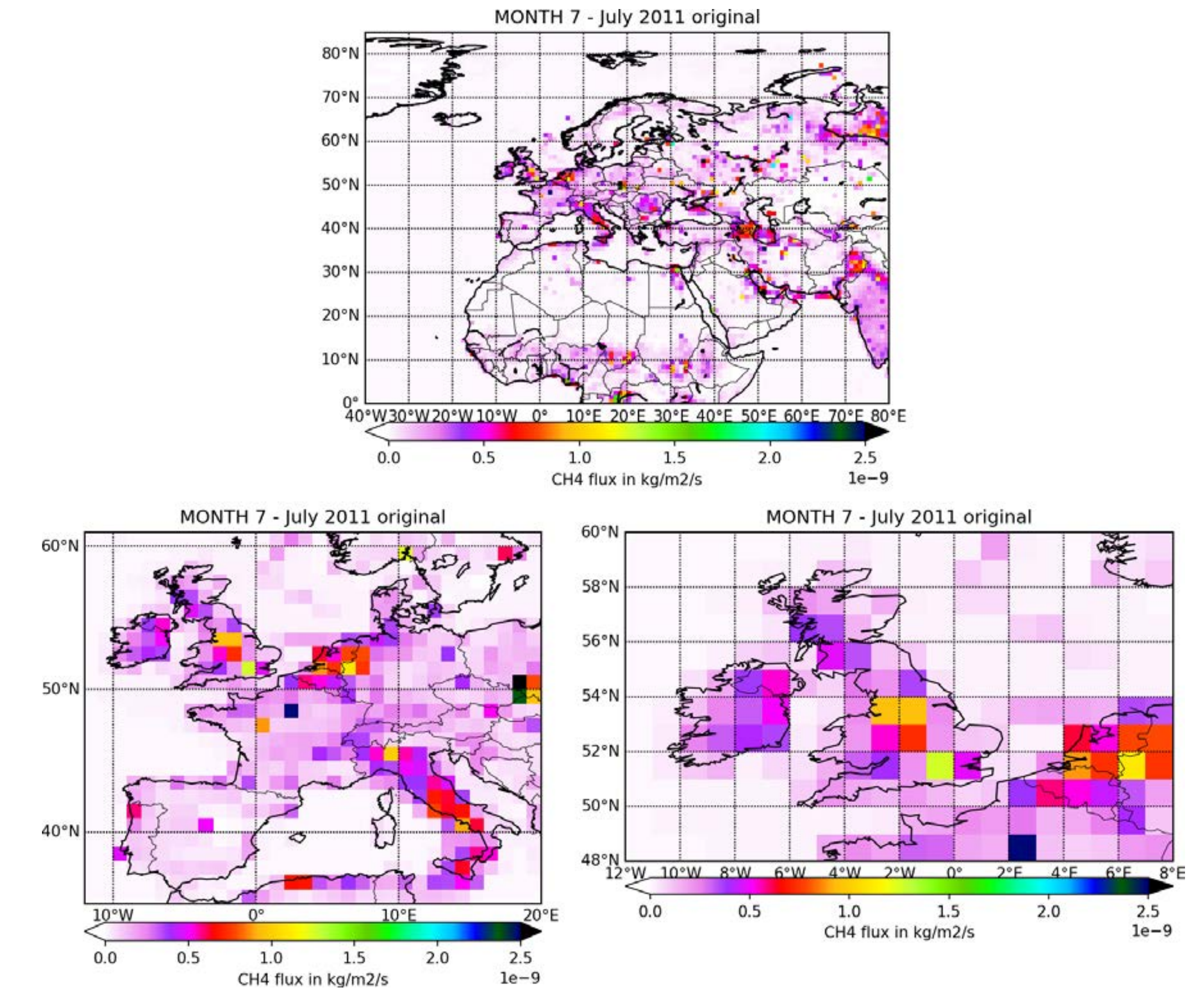

Figure 2.3. Plot of the EDGAR_1.0×1.0_NAT emission inventory, which includes both anthropogenic and natural CH_4 sources, over a wide domain (top left), zoomed into the European domain (top right) and further zoomed into Ireland and Britain (bottom).

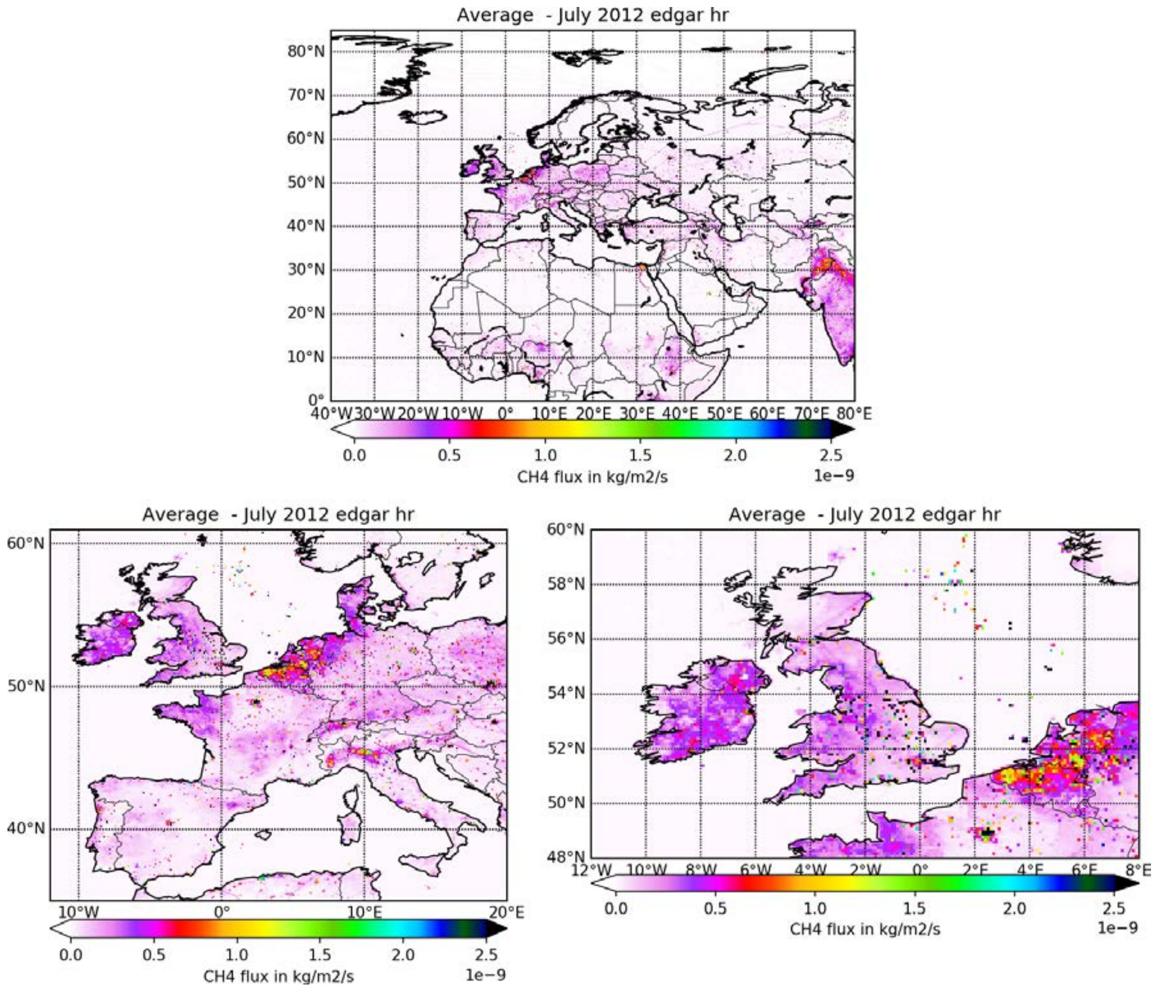

Figure 2.4. Plot of the EDGAR_0.1×0.1_ANT emission inventory, which includes only anthropogenic sources, over a wide domain (top left), zoomed into the European domain (top right) and further zoomed into Ireland and Britain (bottom).

Finally, the total error covariance matrix for the fluxes is the product of the correlation matrix and the error covariance of the a priori fluxes. This requires the calculation of the error on the a priori flux in each grid cell. FLEXINVERT calculates such errors as a fraction of the maximum of the eight surrounding grid cells, compared with the minimum and maximum a priori errors as specified by the user. Finally, the error covariance matrix is scaled by an error fraction of the whole inversion domain. All of these parameters are set by the user in the control file.

Background concentrations (Thompson *et al*., 2015) for 2012 were based on the NOAA flask data; for the higher levels, the Topographic Mapping (TM5) data from the Monitoring Atmospheric Composition and Climate (MACC) project were kindly provided by Rona Thompson.

FLEXINVERT either can use data as described above as background mixing ratios or, alternatively, can be based on observational data after a smoothing process. Although the option of choice in the IMPLiCIt project was to use the re-gridded observational and model data provided by Rona Thompson, an initial comparison with the clear air samples of Mace Head station was performed to understand whether or not the background values used for the Irish domain are actually in line. A small domain such as Ireland may be more sensitive to the background selection and, if the background is too low, the inversion will compensate for this and generate a larger emission flux. Figure 2.7 shows the Mace Head background smoothed (with a moving average window taking only the values below the 50th percentile) time series (orange line), with the background mixing ratio used in the inversion shown by the yellow line.

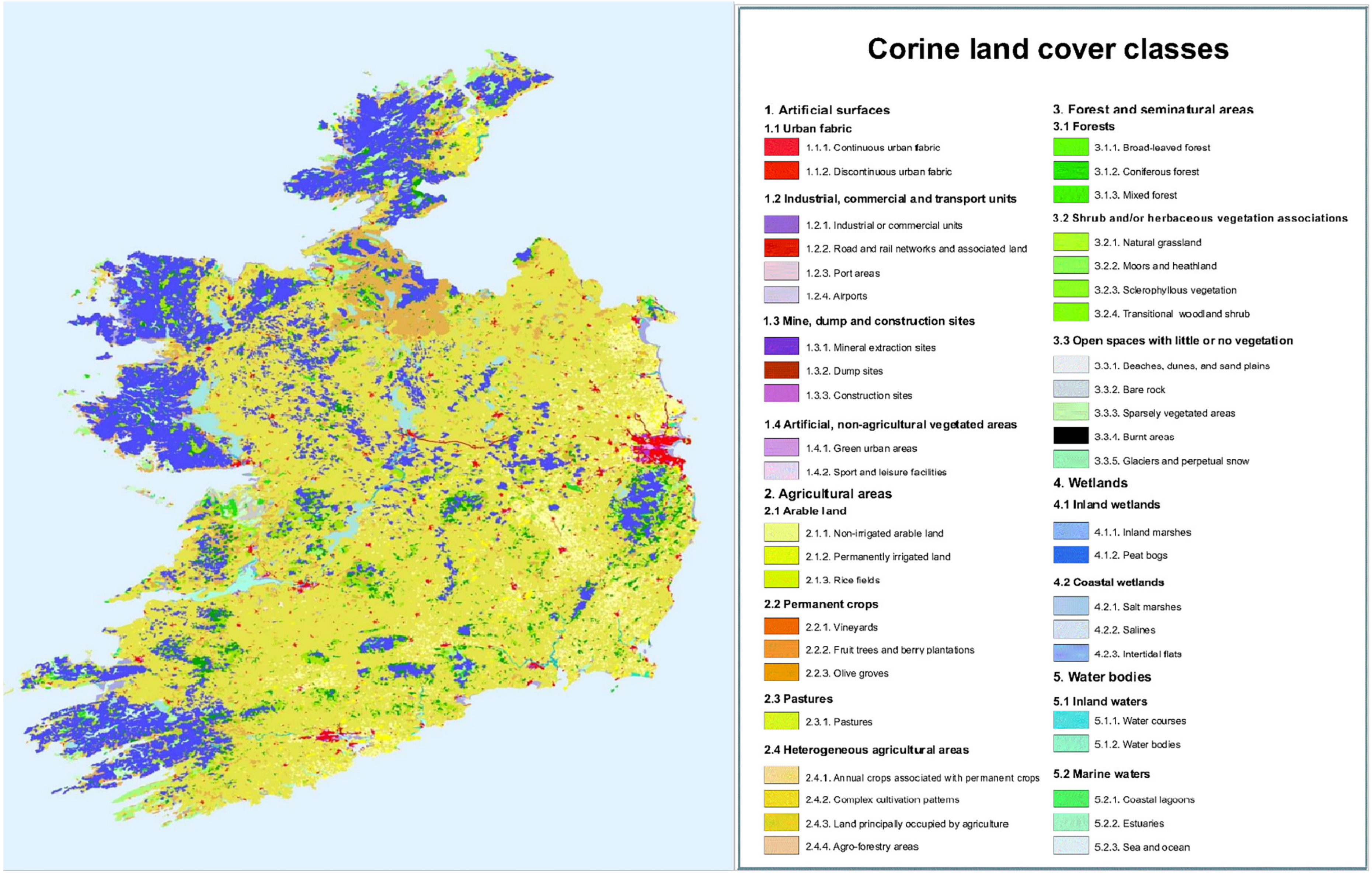

Figure 2.5. Plot of the CORINE land cover types of 2012. Source: Lydon and Smith (2014).

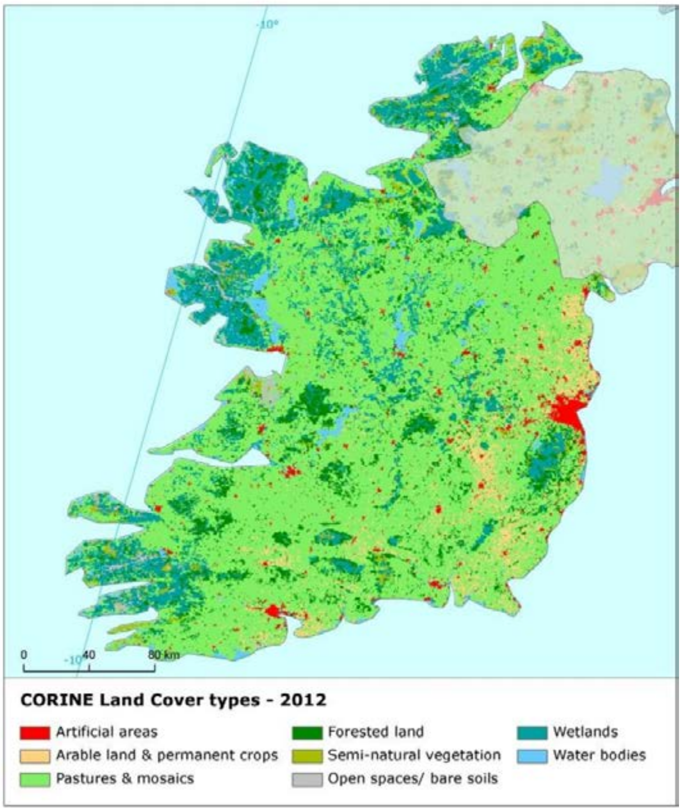

Figure 2.6. Simplified plot of the CORINE land cover types of 2012. Source: EC (2017).

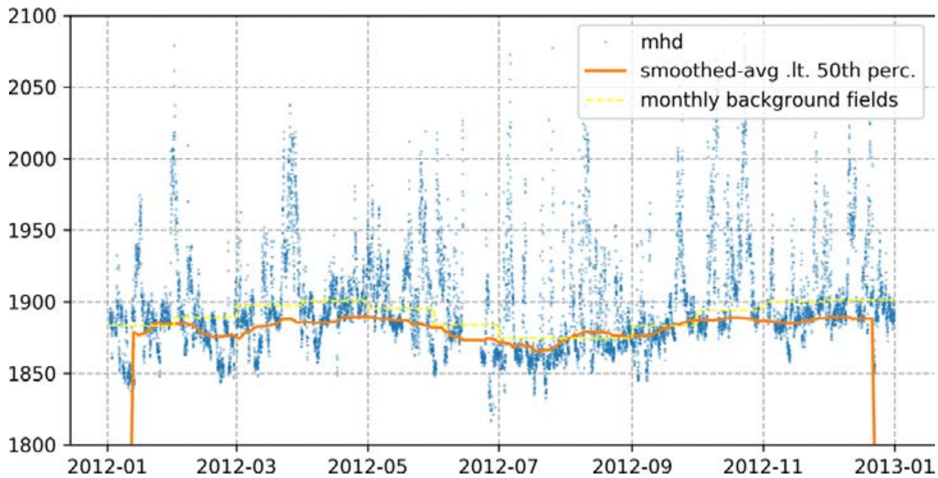

Figure 2.7. Mace Head time series (blue), background derived (orange) and background used in the inversion (yellow).

2.4 Atmospheric Transport Modelling

2.4.1 The FLEXPART model

The FLEXPART model is a Lagrangian particle dispersion model (LPDM) that calculates trajectories from a large number of computational particles. These trajectories include not only the transport resulting from the mean wind (as in the traditional trajectory models) but also the transport component due to turbulent motions occurring at the sub-grid scale, as well as the mesoscale fluctuations (meandering) and deep convection.

In addition, the FLEXPART model includes deposition processes, both dry and wet, gravitational settling for aerosol particles, radioactive decay (also applied to the deposited fields) and a basic linear chemical reaction with the OH radical. This is of particular interest to the IMPLiCIt project as the loss process of CH_4 through the reaction with the OH radical should also be taken into account. FLEXPART is not a full chemical transport model (CTM) and hence does not include full chemistry for the simulated species. FLEXPART is an offline model that uses meteorological fields (analyses or forecasts) as input. Such data are available from several different numerical weather prediction (NWP) models, from the IFS (Integrated Forecast System), the European Centre for Medium-Range Weather Forecasts (ECMWF), the GFS (Global Forecast System) and the US National Centers for Environmental Prediction (NCEP). Other FLEXPART model branches have been developed for input data from various limited-area models, for example the Weather Research and Forecasting (WRF) meteorological model or the Consortium for Small-scale Modeling (COSMO) model.

One of the main advantages of LPDMs such as FLEXPART is that they are naturally self-adjoint. This means that they can run easily both forwards and backwards in time from either the source or receptor locations (Seibert and Frank, 2004), without having to have a specific, and usually fully up-to-date, adjoint version of the model. Some of the advantages of LPDMs compared with the Eulerian CTMs are as follows:

- there is limited numerical diffusion because of the Lagrangian framework;
- better simulation of point measurements/ emissions as the particles concentration not immediately diluted in a computational grid cell;
- better representation of narrow plumes or filaments;
- multiple species can be transported in the same computational particle, preventing the computational increase associated with an increase in the number of pollutants considered.

On the contrary, LPDMs, and FLEXPART in particular, have the following disadvantages:

- There are potential uncertainties from interpolation of the mean fields coming from the meteorological driving data into the particle positions.
- Although the computational demands of LPDMs are normally lower than those of Eulerian models, they scale approximately linearly with the number of tracked particles, and therefore trade-offs between the number of particles and computational resources need to be managed.
- There are uncertainties associated with the number of particles. A small amount of particles will lead to larger uncertainties; this is important for longer simulations in which the particles are gradually spread into a wider area. In brief, the statistical error in the output decreases with the square root of the particle density and this means that the error will increase with distance to the source.
- Limited (linearised) chemistry.

Because of their many advantages, LPDMs have been used in a wide variety of applications, including inverse modelling of trace substances, nuclear emissions and GHGs. Since its very start, in 1998, FLEXPART has been used in an increasing number of applications, from its original scope, specifically for nuclear releases on behalf of the NBC (Nuclear, Biological, Chemical) defence unit of the Austrian military, to the present, with applications worldwide, ranging from pure research to even operational schemes such as that of the Comprehensive Nuclear-Test-Ban Treaty Organization or the Austrian Weather Service.

2.4.2 Model set-up

The FLEXINVERT model was initially developed for global applications and it is therefore internally configured to work with global fields for the atmospheric transport modelling calculations of the source receptor relationships. However, an offset branch to work with non-global SRS fields (actually, to use global SRS fields with a nested domain at a higher horizontal resolution) was developed in a different project and used in the IMPLiCIt project. This facilitated the ability to test different resolutions in the atmospheric transport modelling calculations. It is important to note that the higher the resolution, the larger the computational demands for running FLEXPART. For instance, the executable can easily reach 40 GB if the FLEXPART resolution is 0.2 and the code is driven with meteorological data with 0.5 degrees in a global domain and 0.125 degrees in a nested domain. This computational expense largely constrained the maximum resolution achievable in the IMPLiCIt project.

Source receptor sensitivity files at two different resolutions for the year 2012 were generated. A third set of files was generated with the new version of FLEXPART needed to run FLEXINVERT+. The higher resolved domains are nested into a global 1 × 1 degree domain and the sizes differ as detailed in Figure 2.8.

These calculations have the following technical details and specifications:

- FLEXPART version – two versions were used: version 9, with an adapted version of the OH reactions, and version 10 beta for FLEXINVERT+
- Meteorological driving data – the analyses for the ECMWF were used. As it is possible to use nested driving data with FLEXPART, two domains of meteorological data at different resolutions were used: (1) a global domain at 1 × 1 degree horizontal resolution and (2) a constrained domain over Europe with a 0.125 × 0.125 degree horizontal resolution. Both sets of data have a time resolution of 3 hours.
- Horizontal resolution of FLEXPART output domain(s) (SRS) – the SRS files were calculated in a global domain at 1 degree horizontal resolution, with nested domains at 0.5, 0.2 and 0.125 degrees.
- Temporal resolution of FLEXPART output domain(s) (SRS) – daily average output.
- Release specifications:
  - simulated species: CH_4, with the loss of CH_4 through the interaction with the OH radical activated;
  - emissions in 3-hour intervals;
  - particles backtracked for a period of 10 days;
  - 50,000 particles released at every interval.

2.4.3 Source receptor sensitivity calculations

Using the defined specifications, different sets of SRSs were calculated. FLEXINVERT, unlike its successor version FLEXINVERT+, does not provide the user with the code to generate the large number of runs required in such a study. It was therefore necessary to develop

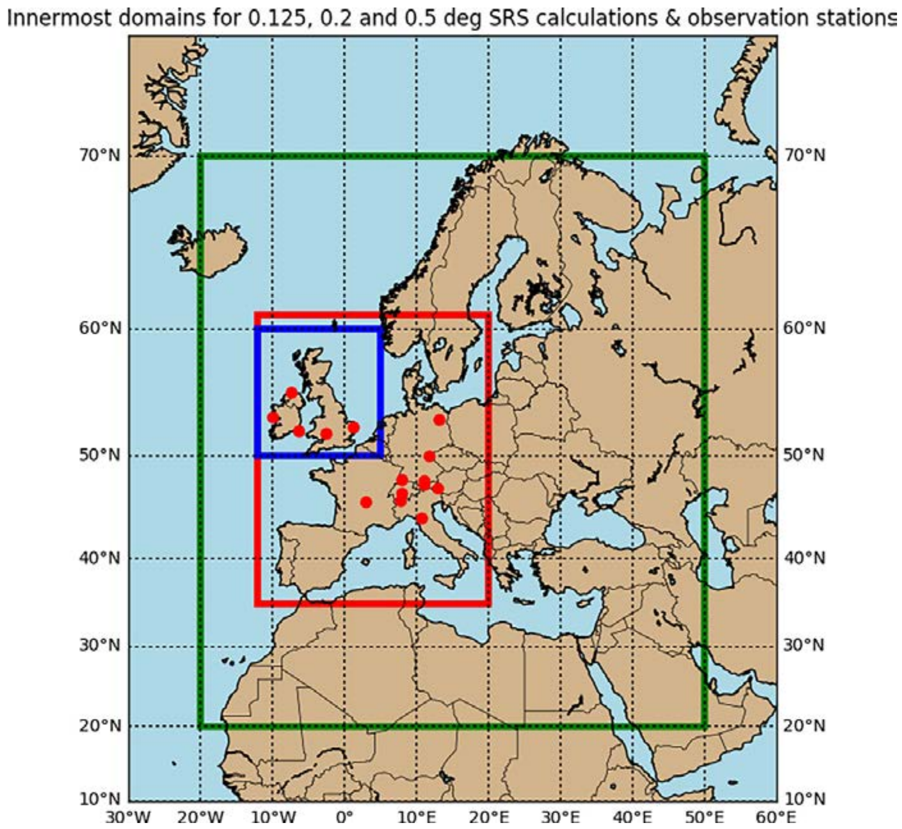

Figure 2.8. Innermost domains used in the SRS calculations, nested into a global 1 × 1 degree domain. Green, 0.5 × 0.5 degrees; red, 0.2 × 0.2 degrees; blue, 0.125 × 0.125 degrees. The blue domain is to be used only in the potential FLEXINVERT+ inversions. The red dots are the observation stations used in the inversions.

specific software to organise the runs while preparing the input data, controlling the output and distributing the runs among processors in a multiprocessor server. A simple Python suite was developed for this task. The suite (Figure 2.9) was developed together with the corresponding required libraries and README and HOWTO documentation.

This code includes:

- The main handling routine, run_flexpart_inversion.py. This routine handles all of the intermediate steps and the unit runs. The user needs only to adapt the first path to make it match the directory where the code is running, that is, the current working directory.
- The static data is in the FLEX_base directory. Here, the user will need to define the OUTGRID, OUTGRID_NEST, AGECLASSE and COMMANS_bloc2.txt files according to the simulation needs.
- sites.txt, to select the sites where the releases should be performed (the measurement sites).
- The controlling file where all of the paths and other general options are specified is namelist.nml. The user needs to adapt the different fields to match the study. In this file the user can define the maximum number of processors to be used.
- Flexlibrary_flexinvert.py is the library, which has multiple functions to generate the files, track the runs, etc.

These runs can be time-consuming. In a Linux Jessie server with 64 GB of RAM (random access memory) and 16 processors, it takes about half a day to simulate a full year and one site. The output is about 90 MB per run.

It is useful to produce plots for the accumulated SRS (Figure 2.10), which is used in FLEXINVERT to define the variable-resolution grid, with grid cells as small as the FLEXPART nest and as large as 4 × 4 degrees. These plots are useful to identify those areas that the inversion will be able to constrain, based on model output, the emissions – in other words, those areas where there is useful and pertinent information. From the images in Figure 2.10 it is evident that using only the three Irish sites will constrain, at most, the Irish domain; however, additional sites would be advisable (note that the colour is more towards the blue in the north and centre in Figure 2.10).

The quantification of the uncertainty associated with the SRS fields (basically, with any atmospheric transport model simulation) is complex and has been discussed at length in the research community (e.g. Seibert, 2000; Hanna *et al*., 2007; Mathieu *et al*., 2018). It is hard to account for and quantify all of the sources of uncertainty involved, which include both interpolation and numerical errors, physical parameterisations (often based on a limited and/or idealised observational dataset), advection schemes and the uncertainties associated with the meteorological driving data, which can be large. Given the large unknowns in the uncertainties, it is difficult to approach the quantification of error through a formal error propagation method. In FLEXINVERT, the errors associated with the model are extracted from the FLEXPART run. They account only for the statistical errors resulting from the number of particles per grid cell. This is, of course, quite a limited approach. Ideally, a Monte Carlo approach should be used so that an ensemble of source terms can be produced based on a perturbation. The new version

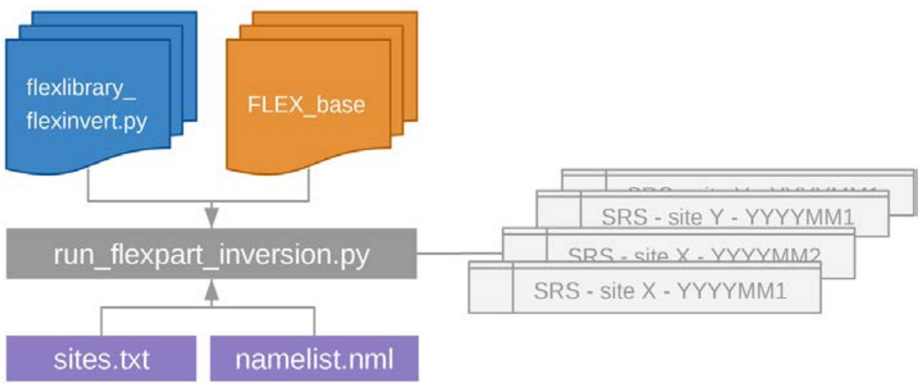

Figure 2.9. Diagram of the structure and main files needed to run the unit runs to generate the SRS files automatically.

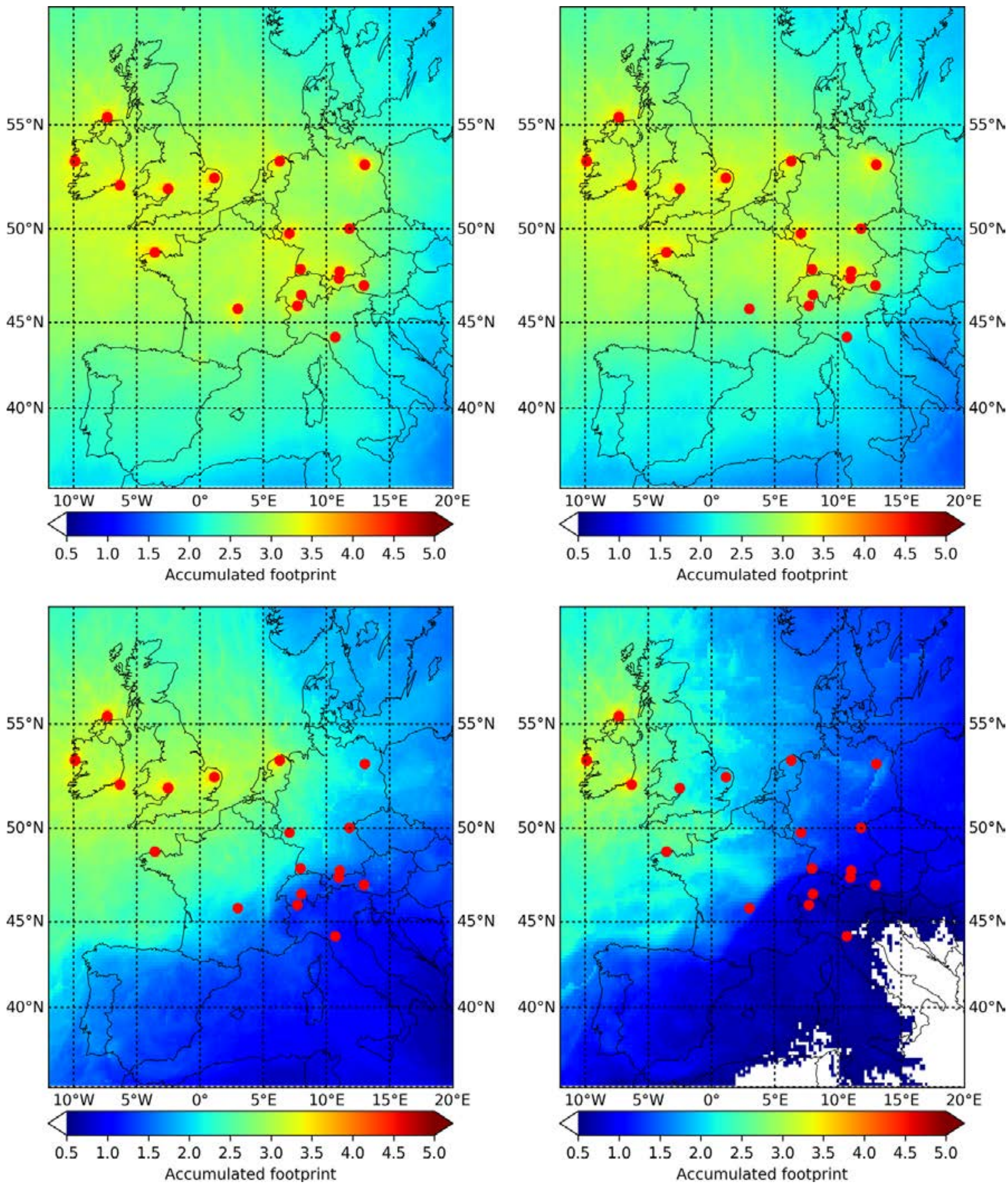

Figure 2.10. The 2012 accumulated footprints in log10 (sec) for all sites (upper left panel), the subset_ sites set (upper right panel), the ie_uk_sites (lower left panel) and just the Irish sites (lower right panel).

of FLEXINVERT, FLEXINVERT+, includes this option and should be investigated in follow-up activities. Alternatively, the errors in the atmospheric transport modelling could be pragmatically obtained in the usual ensemble approach whereby multiple atmospheric runs would be to obtain, per grid cell, a range of values that would account (partly) for the uncertainty. Driving the model with different input data would, for instance, allow a first estimate of the uncertainty associated with the meteorological fields used in the calculations.

3 Evaluation

An initial qualitative evaluation was performed by visually comparing the two-dimensional maps and difference fields of the flux in the different domains against each other and against the a priori information and MapEIre results. Time series at the observational sites were also visualised with the observed measurements and the simulated measurements (those obtained by folding the SRS fields with the estimated emissions inventory). This qualitative evaluation was supported by the evaluation of the statistical scores of correlation (*R*), root mean square errors (RMSEs) and normalised standard deviations (NSDs) at selected stations.

3.1 Comparison with Independent Station Data

Different sets of observational data were used in the inversions. It is clear that, for proper evaluation of any results, independent data should be used. Given the availability of station data for the year 2012, the following two approaches were used:

1. Take the ie_uk_sites runs and compare the modelled time series at the remaining sites. This, however, has the inconvenience that we will be evaluating the European estimated flux at sites in southern Europe or central Europe, whereas we are actually interested in the results for the Irish domain. Although this limitation is evident, this approach still provides some assessment of the general performance of FLEXINVERT for this study.
2. Use the all_sites runs but perform the inversion calculations using only a subset of data for the three Irish sites. This allows us to use the remaining data as an evaluation system, while focusing on our region of interest. There are several ways to select a subset of observational data from a time series. A random selection would simply take random observations out from the whole period. However, it is worth noting that we do expect to have some temporal correlation in the measured CH_4 according to background and weather patterns. Without making a detailed study it can be argued that an approximate time window of about 4 days would prevent the taking of correlated data on many occasions. Therefore, a system was programmed that selects the data from the three sites over a 4-day window, with a different start time in each of the sites. Half of the observational data from the Irish sites are used only in the evaluation and not in the inversion.

3.2 Comparison with the 2015 High-resolution Emission Inventory

Unfortunately, a detailed emissions inventory for Ireland for 2012 was not available at the time of this project. However, the national mapping of GHG and non-GHG emission sources project (MapEIre; see http://projects.au.dk/MapEIre/; Figure 3.1), funded by the Irish EPA, has produced a high-resolution emission inventory for CH_4 for 2015 that could be used for evaluation. Its SPREAD (spatial high-resolution distribution model for emissions to air) model results provide shape files that include emissions from the national emissions inventories for air pollution and GHGs, spatially distributed on a grid with a geographical resolution of 1 km × 1 km, using the Irish projection TM65. In addition, information on the sectoral level corresponding to the goods not for resale (GNFR) sectors used for reporting of gridded emissions to the United Nations Economic Commission for Europe (UNECE) Convention on Long-range Transboundary Air Pollution (CLRTAP) is provided, without having the land use, land use change and forestry (LULUCF) sector included in the GNFR sectors. Discussion with members of the MapEIre project has facilitated, on one hand, access to the data and, on the other hand, the required information to convert from the Irish TM65 grid to the WGS89 (World Geodetic System) latitude/longitude data. With this information, a re-gridding routine has been prepared that allows, first, re-gridding into a regular latitude/longitude grid and, second, downscaling to the desired resolution, keeping the total emissions as close as possible to the original emissions (from 545.42 kt to 551 kt) so that the results can be compared with the estimated fluxes. National

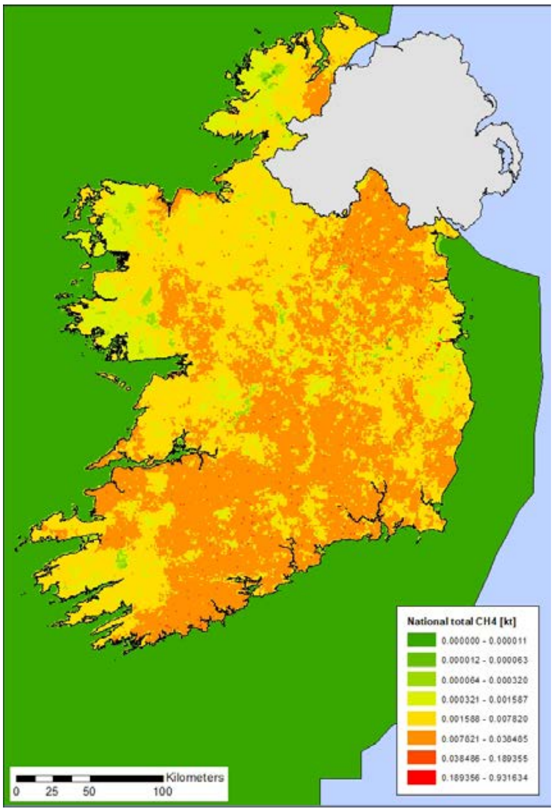

Figure 3.1. High-resolution CH_4 emission inventory, as generated in the MapElre project. Source: Nielsen *et al.* (2019).

total CH_4 emissions in 2012, as reported by the EPA in 2017 (Duffy *et al.*, 2017), were 502.84 Gg, including LULUCF and memo items. For 2015, as estimated in the MapElre study, the total emissions were 545.42 Gg. That is a factor 1.08 difference between the 2 years.

As previously specified, two emissions inventories were used, EDGAR_1.0×1.0_NAT and EDGAR_0.1×0.1_ANT, that, in the re-gridded space, sum to 671 Gg and 659 Gg respectively.

Figure 3.2 shows a visual comparison of the inventories.

To perform the evaluation, focus on the Irish domain was required. To achieve this aim, a system to mask out the emissions was needed so that emissions from outside Irish national boundaries were not accounted for. A simple Python code was developed that simply generates a gridded set of 0 or 1 depending on whether the points are inside or outside Irish political boundaries. Once this 0–1 mask is generated, it can be easily folded with emissions inventories to constrain the region of interest.

3.3 Comparison with Similar Studies

A limited number of studies have looked specifically at Irish emissions. The study by Ganesan *et al.* (2015) suggests that inferred emissions in Ireland were up to 30% higher than the a priori emissions. This is in line with the results presented here, with differences seen between a priori and the FLEXINVERT inferred emissions.

3.4 Comparison with a Different Inversion System

An alternative inversion system was used for comparison in the iMPLiCIt project based on previous experience in other studies. LOGINVERT assumes a log-normal distribution for the observation and a priori errors in the cost function. These assumptions are closer to the distribution of observed concentrations and surface fluxes than Gaussian assumptions (Brioude *et al.*, 2011, 2012). As a consequence, this prevents the posterior estimate having unrealistic negative fluxes. The drawback is that no analytical solution is found for the inverse problem and an

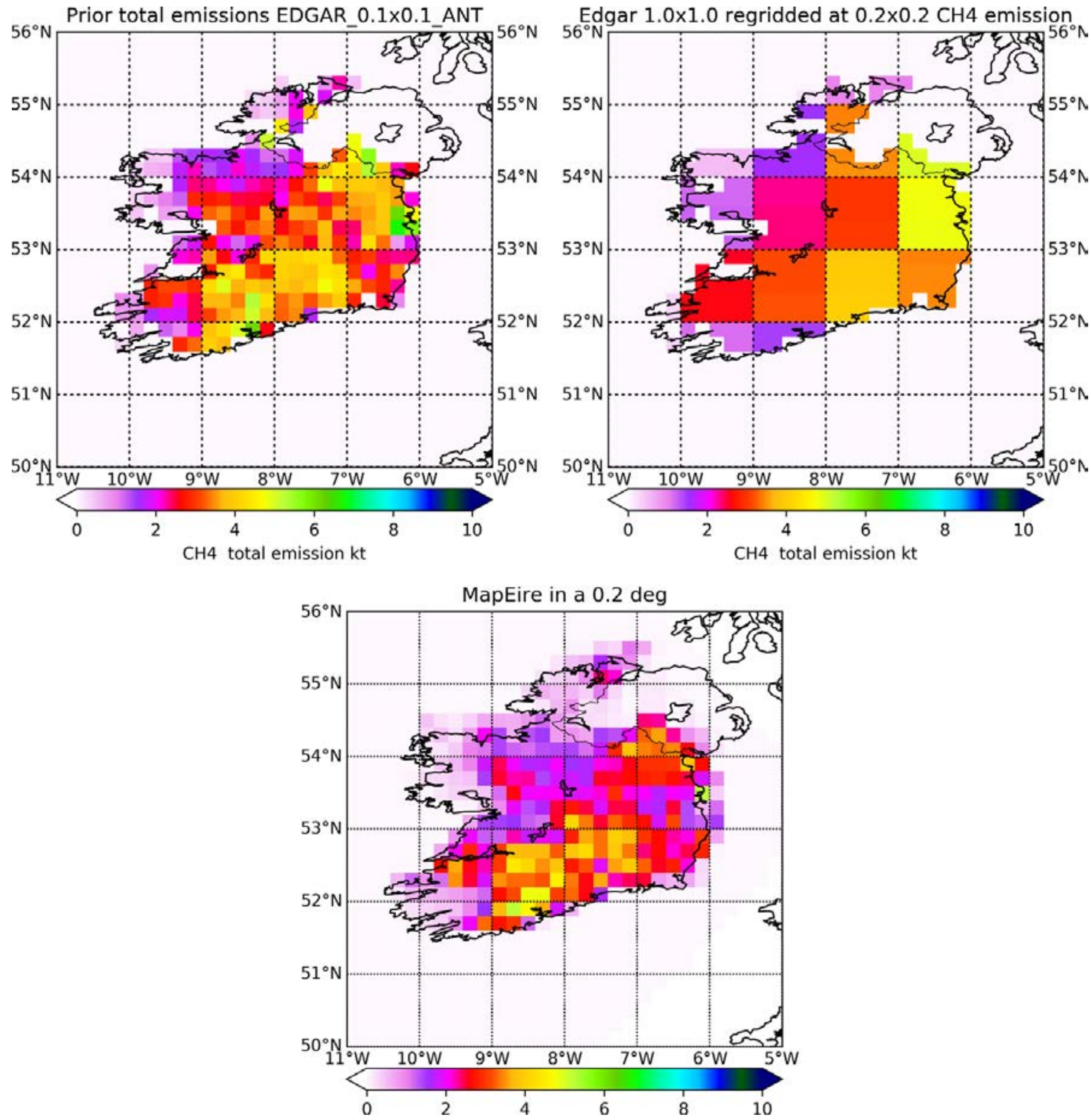

Figure 3.2. EDGAR_0.1×0.1_ANT emission inventory (left) re-gridded into a 0.2 grid (middle) and MapEIre inventory (right) for 2015 also downscaled at 0.2 degree grid with the process described in the text. Note that the MapEIre inventory includes natural sources whereas EDGAR includes only anthropogenic sources.

iterative method needs to be used to converge to a solution. LOGINVERT has been modified to make use of some of the FLEXINVERT output files to facilitate comparison between the model results, given the most similar input and set-up files as possible. Specifically, LOGINVERT will use the same observations, the same background, the same interpolated a priori data and the same SRSs as those used in FLEXINVERT. The covariance matrices obtained from FLEXINVERT were included in the LOGINVERT calculations.

Only two parameters need to be set to run LOGINVERT: the observation and a priori errors. By default, the background uncertainty was assigned a value of 20% (see Angevine *et al*., 2014) and is included in the observation covariance matrix. An uncertainty of 50% was assumed for the a priori errors.

No cross-correlation was assumed in the observation and a priori covariance matrices. The cross-correlation in the observation covariance matrix, which represents the temporal and spatial correlation in the measurements and the model error, is difficult to evaluate without an ensemble of SRSs. The cross-correlation used in the a priori covariance matrix from FLEXINVERT (by default, 100 km over land) can be used in LOGINVERT. However, based on a WRF ensemble (W.M. Angevine, Cooperative Institute for Research in Environmental Sciences, University of Colorado, and Chemical Sciences Division, Earth System Research Laboratory, National Oceanic and Atmospheric Administration, Boulder, CO, 8 January 2018, personal communication) the spatial correlation length scale can vary from 20 to 100 km. By default, LOGINVERT assumes no cross-correlation.

A useful way to increase the likelihood that the observation covariance matrix is diagonal is by taking a random subset of observations so that each observation is not spatially or temporally close to another. This can be done in LOGINVERT. Likewise, to increase the likelihood that the a priori covariance matrix is diagonal, we also applied a random term on the a priori to reduce the cross-correlation between grid cells in the posterior. The random term has the same magnitude flux as the smallest fluxes in the domain. The cross-correlation between grid cells represents the likelihood that a grid cell flux varies owing to the variation of a different grid cell (see Brioude *et al*., 2011 for more details).

Hence, LOGINVERT generates from those random terms around 30 posteriors and takes the mean of the posterior ensembles to obtain the best posterior estimate. A number of cores can be set in the namelist to run LOGINVERT in parallel using a Python multitasking library. LOGINVERT takes about 2–5 minutes to run the posterior ensemble and generate a posterior estimate.

4 Results

4.1 Sensitivity Studies

A first step in inverse modelling studies is to try to identify the optimal set-up of the framework while considering the available data, the models and knowledge about errors. For the IMPLiCIT project, a subset of sensitivity studies was performed that aimed to define the initial starting point for further activities to improve the inverse modelling system specifically for the Irish domain. In the tests presented below, yearly averages or totals are compared.

4.1.1 Influence of horizontal resolution

Two sets of SRS files were generated for the inversions with FLEXINVERT, at 0.5 × 0.5 and 0.2 × 0.2 degrees horizontal resolution. The two resolutions are subsequently referred to as SRS0.5×0.5 and SRS0.2×0.2 respectively. The results are shown for the two emissions inventories available for the Irish domain (Figures 4.1–4.4). There are a number of initial observations:

1. The inequality constraint failed for some months and therefore it was not applied. This is the cause of the gaps in the flux estimates. It is, however, irrelevant for the Irish domain.
2. There is an influence of the a priori used.
3. A hotspot in Poland is always present, both in the inversions and the emissions inventories.
4. The SRS0.2×0.2 files allow for modified fluxes in the south of Ireland.
5. The SRS0.2×0.2 files allow for a larger number of grid cells in the relatively small Irish domain.

Given the size of Ireland, and noting that the fluxes seem to be properly constrained with the observations, leads to the conclusion that using the 0.2 degree resolution SRS (see Figure 4.2) and not the coarser 0.5 degree resolution SRS (see Figure 4.1) is advisable.

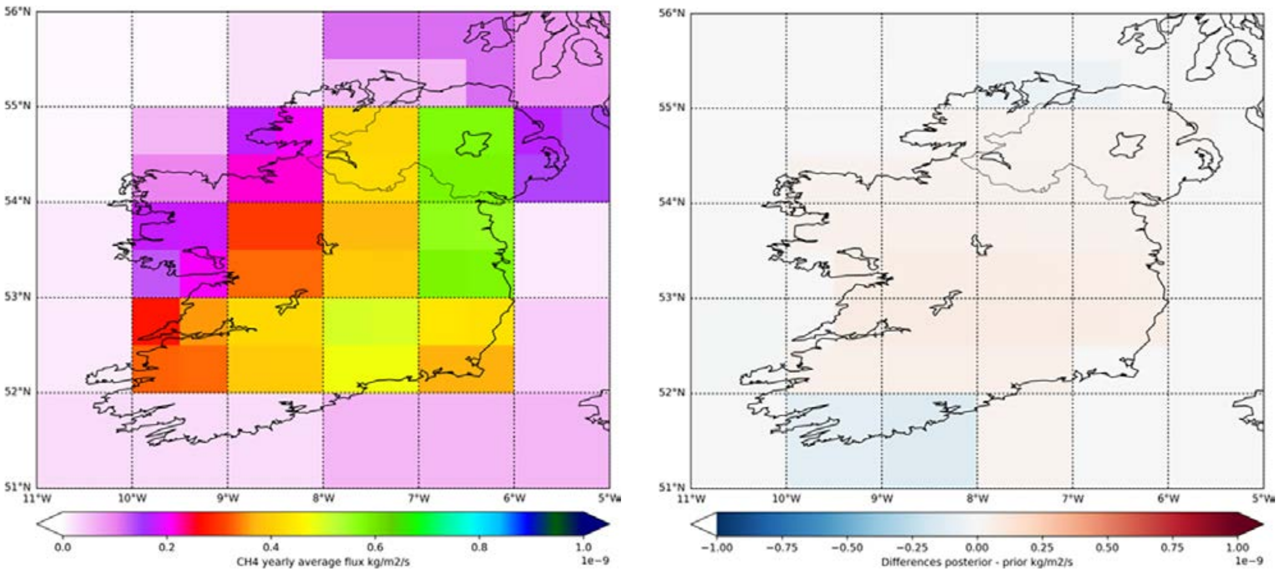

Figure 4.1. Posterior average flux (left) and differences with respect to the a priori (right) for the 0.5 degree SRS files (SRS0.5×0.5) and the 1 degree emission inventory provided by Thompson (EDGAR_1.0×1.0_NAT).

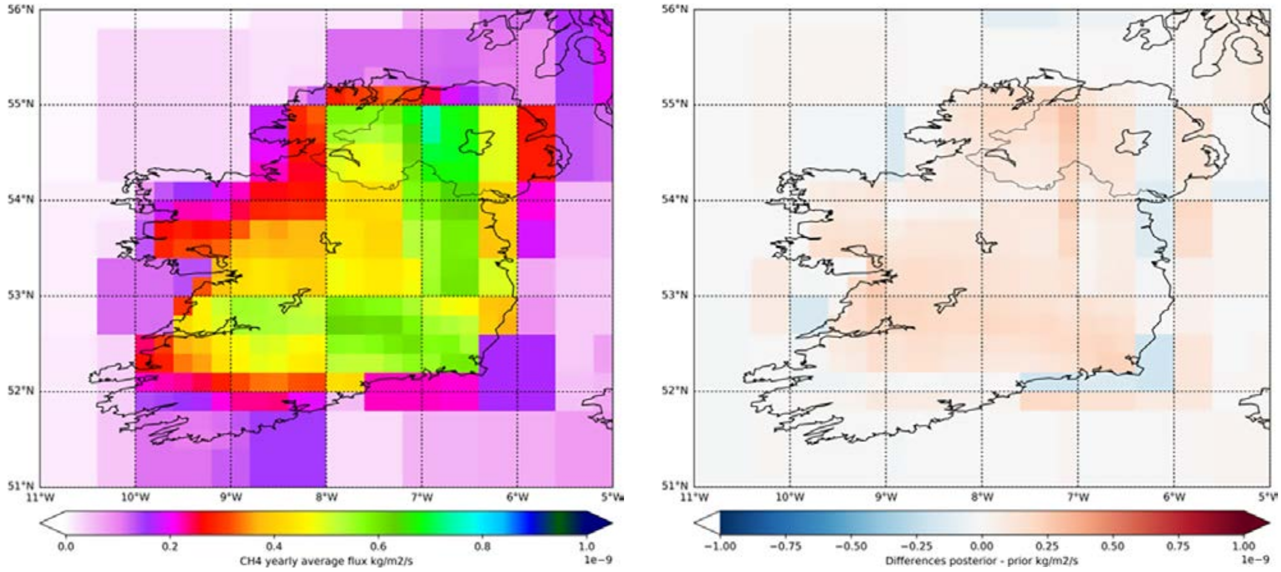

Figure 4.2. Posterior average flux (left) and differences with respect to the a priori (right) for the 0.2 degree SRS files (SRS0.2×0.2) and the 1 degree emission inventory provided by Thompson (EDGAR_1.0×1.0_NAT).

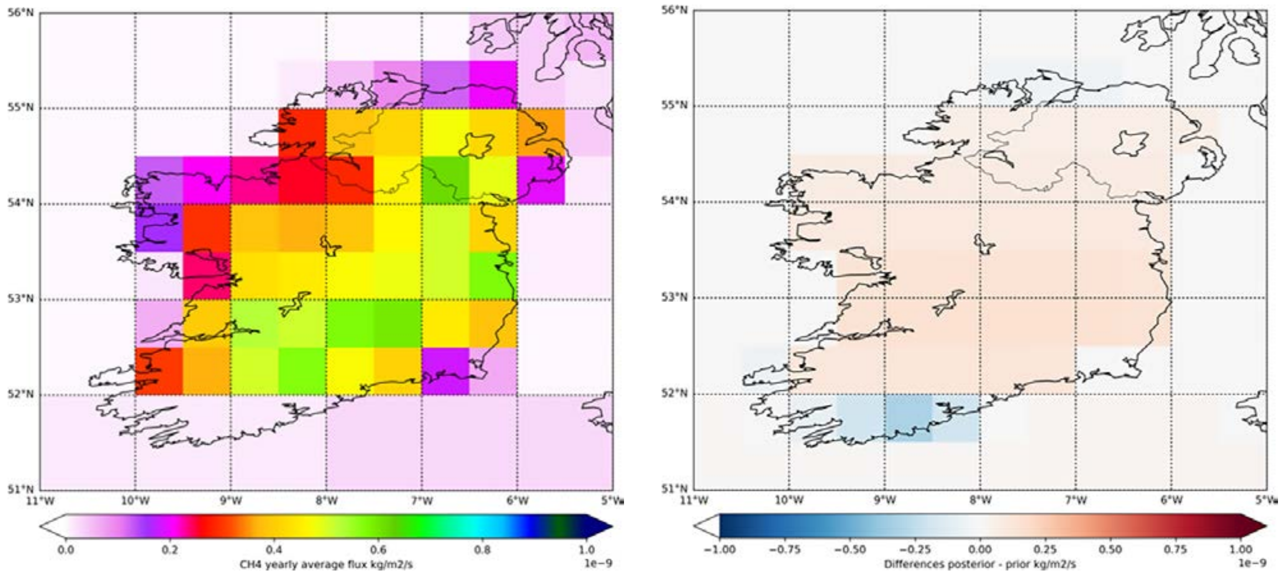

Figure 4.3. Posterior average flux (left) and differences with respect to the a priori (right) for the 0.5 degree SRS files (SRS0.5×0.5) and the 0.1 degree EDGAR emission inventory (EDGAR_0.1×0.1_ANT).

4.1.2 Influence of the a priori

As stated above, two inventories were used, an inventory with a coarse (1 × 1 degree) resolution but including natural emissions from Thompson, and the EDGARv4.3.2 high-resolution inventory, which includes solely anthropogenic emissions. As shown in Figures 4.3 and 4.4, for both inventories and with this set-up, FLEXINVERT significantly increases the emissions in Ireland in eastern and southern areas and Northern Ireland, whereas the north-west remains similar to the a priori used. With the modified 2011 inventory, EDGAR_1.0×1.0_NAT, the total emissions obtained are 871 kt whereas, for the high-resolution inventory, EDGAR_0.1×0.1_ANT, emissions of 952 kt are obtained (8.5% higher).

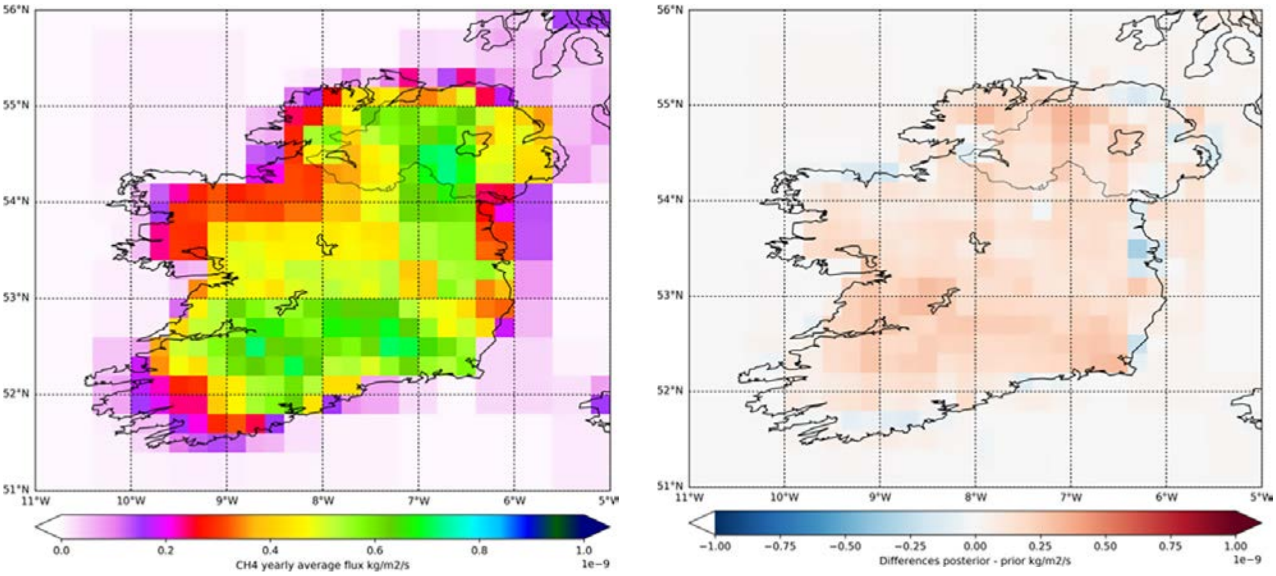

Figure 4.4. Posterior average flux (left) and differences with respect to the a priori (right) for the 0.2 degree SRS files (SRS0.2×0.2) and the 0.1 degree EDGAR emission inventory (EDGAR_0.1×0.1_ANT). Note: the temporal resolution has not been changed as, given that the focus of the IMPLiCIT project is yearly inventories, having daily output for the SRS is enough in all cases; however, for species with a strong diurnal cycle, this may not hold and a different time resolution should be investigated.

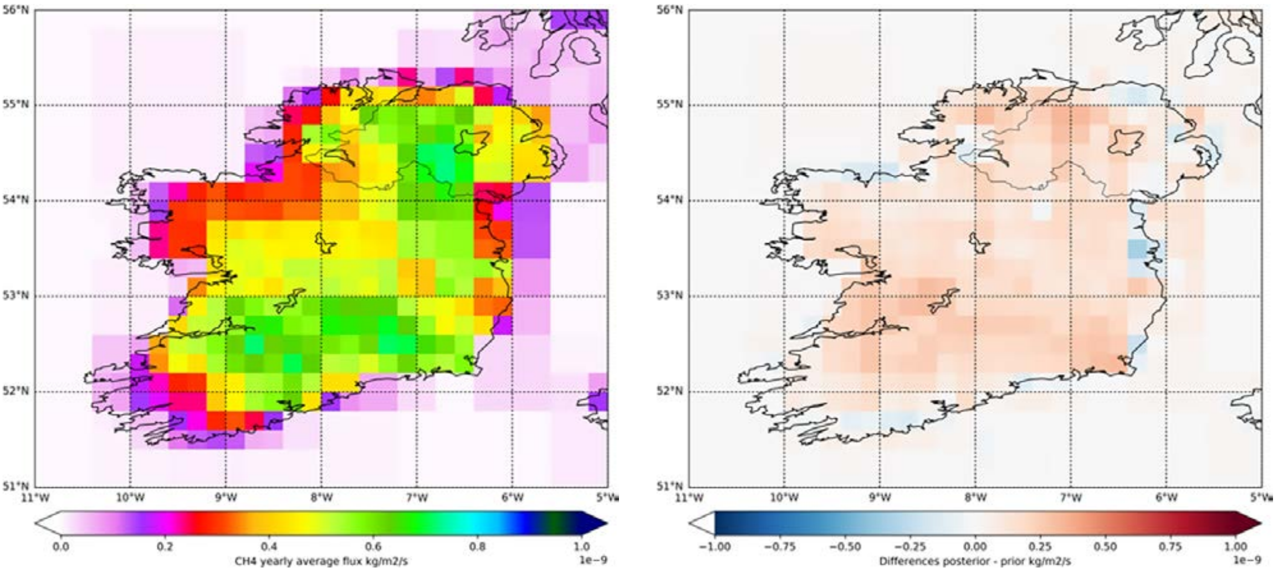

Figure 4.5. Posterior average flux (left) and differences with respect to the a priori (right) for the 0.2 degree SRS files (SRS0.2×0.2) and the EDGAR_0.1×0.1_ANT inventory for the "all_sites" run (estimated total emissions for Ireland are 952 kt).

4.1.3 Influence of the station data used in the inversion

The results outlined for the station data sensitivity test are provided for inversions with the high-resolution emission inventory EDGAR_0.1×0.1_ANT and the 0.2 × 0.2 degree SRS (SRS0.2×0.2; Figures 4.5–4.8).

Comparison of the total inventories when using all of the sites for an inversion (see Figure 4.5) or a subset of sites (see Figure 4.6) shows a relatively small difference, from 952 kt to 940 kt. The main reason for testing a subset of sites was to be able to have independent data for the evaluation. However, given the distance of these stations to the region of interest,

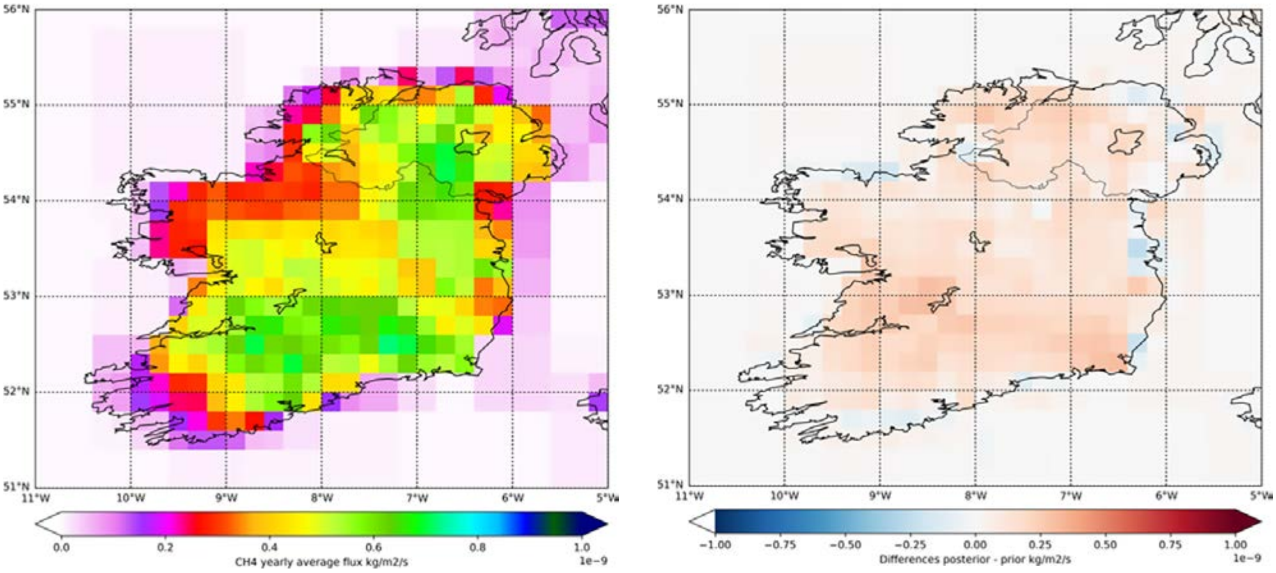

Figure 4.6. Posterior average flux (left) and differences with respect to the a priori (right) for the 0.2 degree SRS files (SRS0.2×0.2) and the EDGAR_0.1×0.1_ANT inventory for the "subset_sites" run (estimated total emissions for Ireland are 940 kt).

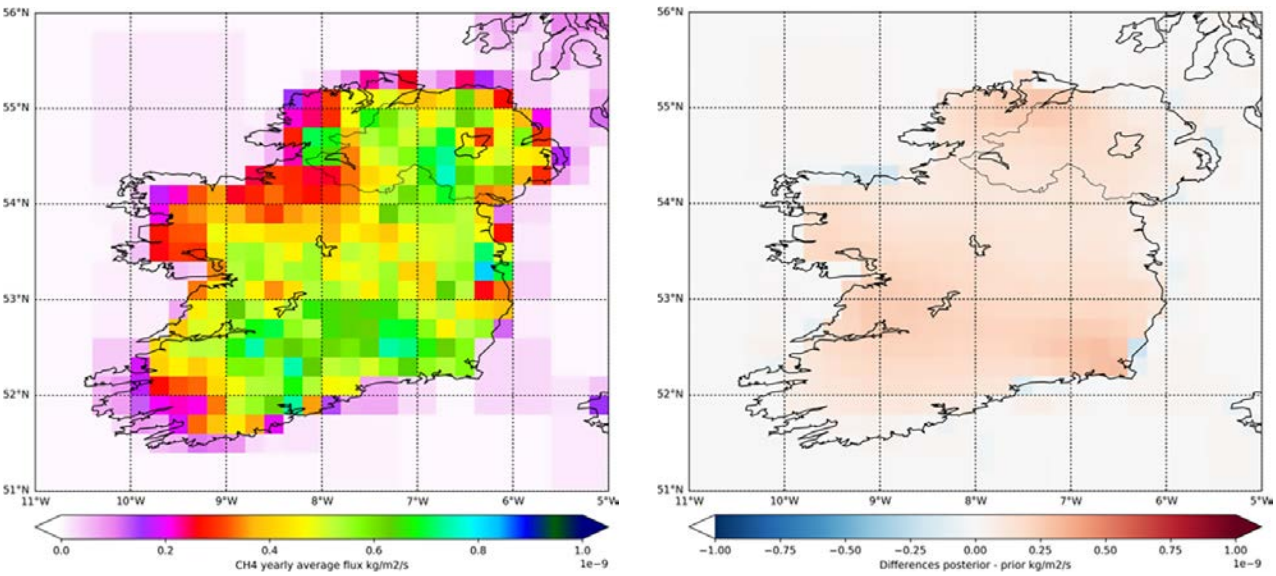

Figure 4.7. Posterior average flux (left) and differences with respect to the a priori (right) for the 0.2 degree SRS files (SRS0.2×0.2) and the EDGAR_0.1×0.1_ANT inventory for the "ie_uk_sites" run (estimated total emissions for Ireland are 977 kt).

it does not seem advisable, and offers no advantage, to use the subset_sites runs.

The inversion was then run using only a small subset of sites comprising those with data available in the UK and Ireland (ie_uk_sites; see Figure 4.7) This does not significantly alter a priori information in southern Europe, where the observational data used in the inversion (or actually the station footprints) are less influencing. The total Irish emissions increase up to 977 kt.

This study had the challenge of having to constrain the emissions of a very small region with only three coastal sites, located in an island with predominant westerly flows and having land only to the east and the south, where the additional measurement stations are. In order to understand how a station may influence the

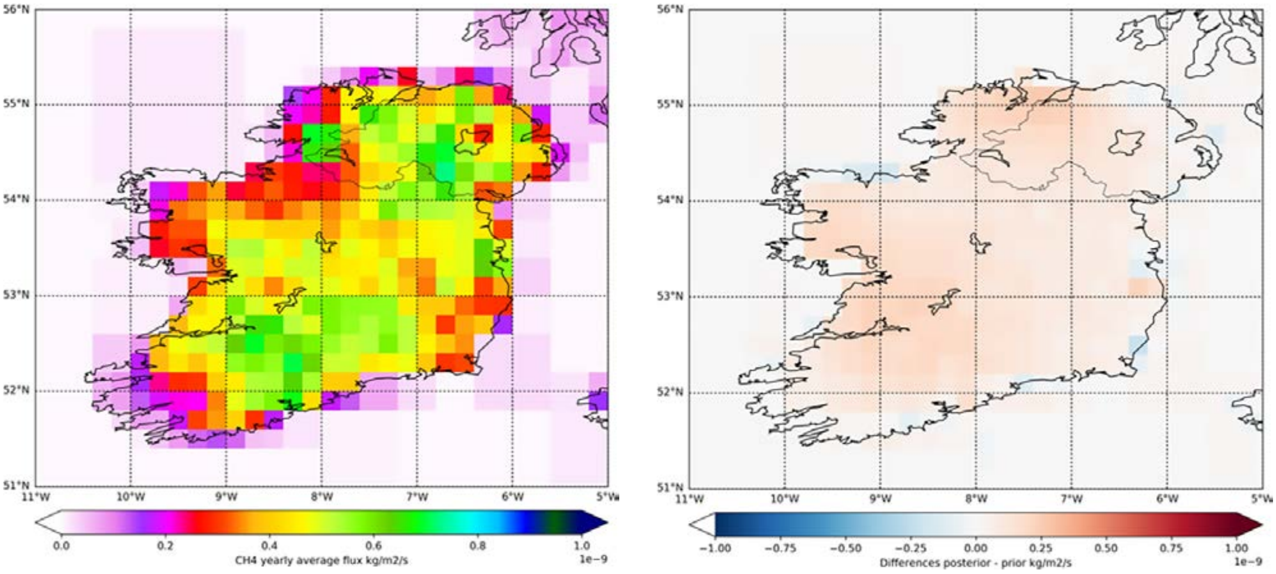

Figure 4.8. Posterior average flux (left) and differences with respect to the a priori (right) for the 0.2 degree SRS files (SRS0.2×0.2) and the EDGAR_0.1×0.1_ANT inventory for the "ie_uk_withoutcarnsore_sites" run (estimated total emissions for Ireland are 895 kt).

posterior fluxes, Carnsore Point was excluded from the ie_uk_sites set (see Figure 4.8).

The results of this test indicate what we might expect: in those areas of influence of stations that are not considered in the inversion, the a priori dominates and the estimated flux tends to the a priori inventory. For the Irish domain, performing the inversion with all of the stations as opposed to the subset_sites makes practically no difference to the total emissions (952 kt and 940 kt, respectively, a difference of less than 1.5%) estimated. Figures 4.7 and 4.8 also exhibit a similar geographical distribution. When only the UK and Irish sites are considered in the inversions, the flux is slightly modified (by about 2.6%), whereas when Carnsore Point station is not used, the overall flux in Ireland decreases (by about 6%), especially, as expected, in the vicinity of the station. In these two runs, it can be seen how the flux in southern Europe matches closely the a priori information because of the lack of influence of the UK and Irish sites used in those areas.

4.1.4 Influence of the observational time window

As shown in the previous sections, irrespective of the set-up, the posterior estimate is larger than the a priori. For these inversions, no selection of the measurements with respect with time was carried out. For stations with strong local emissions, if night-time inversions are included, this may lead to an overestimation of the emissions inventory. To investigate this possibility, inversions have been implemented with only the average of afternoon observations for flat terrain sites and the average of night-time observations for mountain stations (Figure 4.9). This sensitivity case shows no large influence in terms of total estimated fluxes, with a variation smaller than 1% in total.

4.1.5 Influence of correlation length

The observational spatial correlation length is the appropriate length scale used for the simulations and is also a parameter that can be modified. A correlation length of 500 km has been used previously in published FLEXINVERT tests (Thompson and Stohl, 2014). Tests with correlation lengths of 100 km and 500 km were performed and show some differences, affecting the overall total flux for the Irish domain (Figure 4.10). The total emissions calculated using the higher correlation length increases the inferred emissions by approximately 4%.

4.1.6 Influence of the error definition

In the definition of the inversion set-up, uncertainties can be associated with the different parameters. The balance among these will constrain the results more towards the observations or the a priori. In this section,

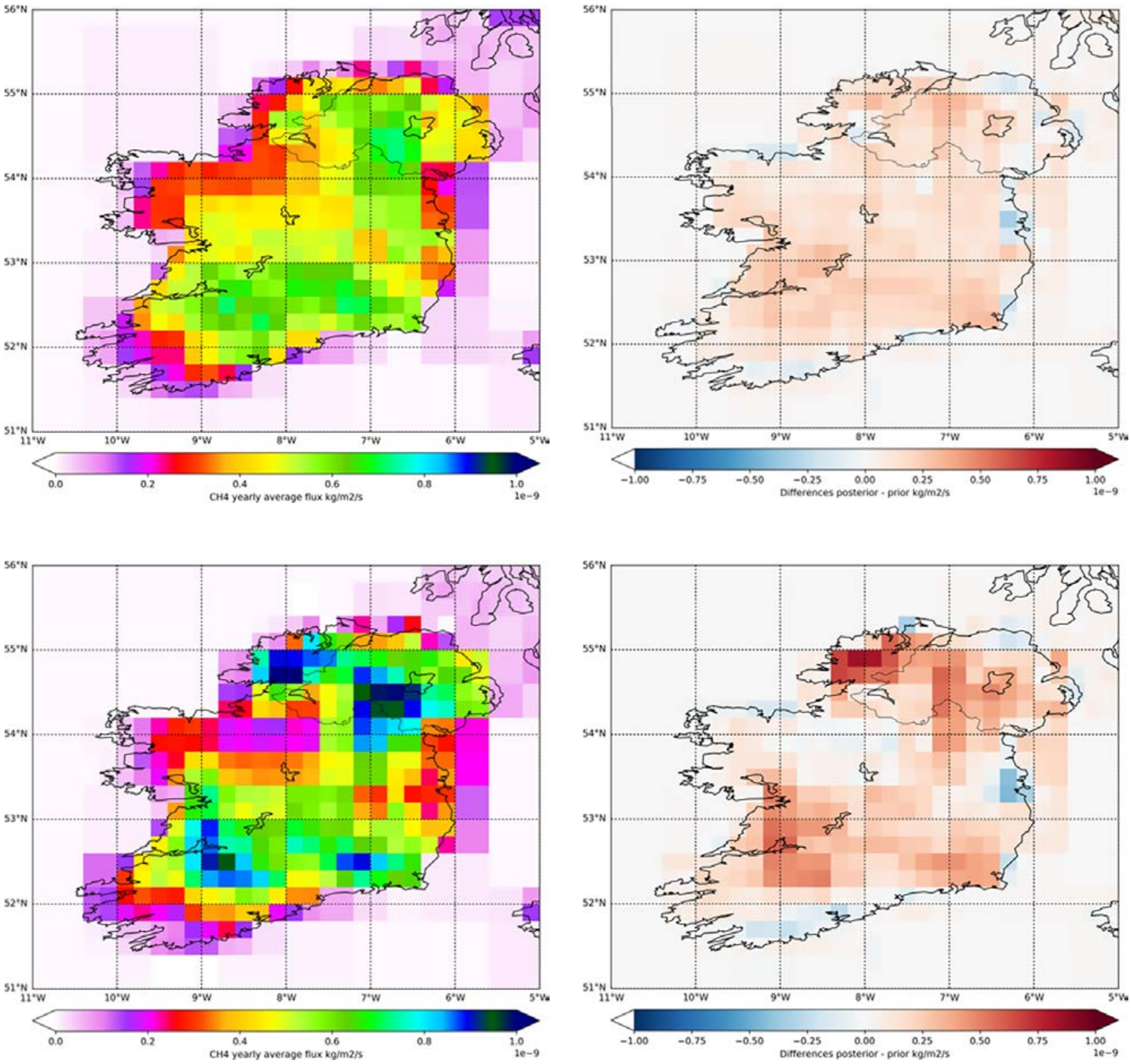

Figure 4.9. Posterior average flux (left) and differences with respect to the a priori (right) for the 0.2 degree SRS files (SRS0.2×0.2) and the 0.1 degree EDGAR emission inventory (EDGAR_0.1×0.1_ANT) with all of the observations (top) and only the afternoon window (bottom) for non-mountain stations.

how the description of uncertainties in the a priori and global inventory affects the results is investigated. As a background, in FLEXINVERT, the a priori uncertainty is calculated as a fraction of the a priori flux. This is carried out for each grid cell but, to not be too sensitive to the a priori flux distribution, the eight neighbouring grid cells plus the one of interest are used to determine the maximum uncertainty. The uncertainties are set to the minimum/maximum values. The error covariance matrix is then scaled to the "global" error set in the file, control.def, where the user sets the different parameters. The selection of the parameters is based on the data distribution, flux estimates and estimated global emissions.

In terms of total emissions (Table 4.1), tests 1 and 2 yield the highest total emissions for Ireland, whereas the basic run and test 4 provide very similar results. Test 3 provides the lowest inferred emissions. However, the variation among all of the runs is smaller than 130 kt.

Looking at the time series for the Irish sites, relatively small differences can be observed among the tests. However, larger differences are observed between inversion systems. It is evident that FLEXINVERT is much more closely constrained by the a priori compared with the behaviour of LOGINVERT, which allows more adaptation to the actual observations.

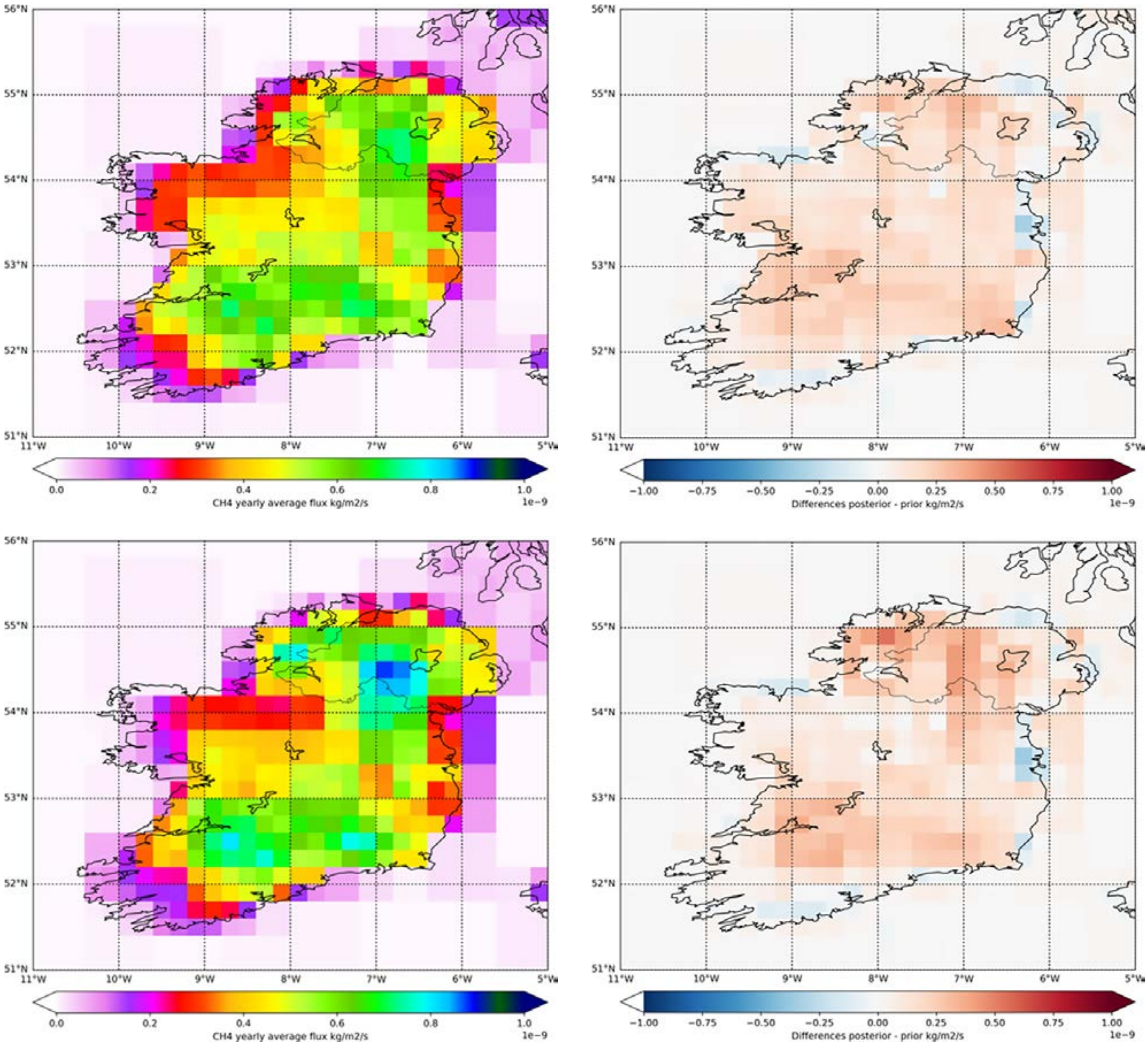

Figure 4.10. Posterior average flux (left) and difference with the a priori (right) for the 0.2 degree SRS files (SRS0.2×0.2) and the 0.1 degree EDGAR emission inventory (EDGAR_0.1×0.1_ANT) showing an afternoon observation window and correlation length of 100 km (upper panels) and an afternoon observation window and correlation length of 500 km (lower panels).

Table 4.1. Description of the tests used to investigate the influence of the error definition

Test name	Min. error in a priori (kg/m²/h)	Max. error in a priori (kg/m²/h)	Global error (Tg/y)	Obs. in unit of observations (e.g. ppb)
Basic	0	1.0×10^{-6}	4×10^{0}	2×10^{0}
Test 1	1.0×10^{-8}	1.0×10^{-2}	10×10^{0}	2×10^{0}
Test 2	1.0×10^{-8}	1.0×10^{-2}	10×10^{0}	5×10^{0}
Test 3	1.0×10^{-8}	1.0×10^{-4}	4×10^{0}	2×10^{0}
Test 4	1.0×10^{-1}	1.0×10^{0}	8×10^{0}	5×10^{0}

However, this comes at the cost of producing higher total emissions for Ireland. It is considered that the LOGINVERT estimates may be unrealistic and largely affected by the uncertainty in the atmospheric transport modelling because of the relatively small number of observations in the region of interest. Of course, the same aspects affect the FLEXINVERT results, also potentially leading to total estimated fluxes that are too high. Figures 4.11–4.15 show the posterior average flux and differences with respect to the a priori for the basis and test runs as well as time series for the different tests and inversion methodologies for the Irish sites. Statistical scores for these time series are shown in Table 4.2. A slight improvement in the scores can be seen when using the flux estimated with FLEXINVERT, the differences are not large enough to consider them a significant improvement. LOGINVERT, however, shows a larger improvement in the scores, which is also shown in the time series, where the maxima are better matched in those simulated values.

In summary, for FLEXINVERT, it does not appear that variations in the quantification of errors significantly modify the scores at the sites and, in addition, the total emissions show a maximum variation of 7%.

On average, the variations among tests (shown in Figure 4.16) are relatively minor and quite similar when only the three Irish sites are considered in the statistics calculations. Based on this, and considering that test 4 is more reflective of the errors, this is the set-up that we consider the most appropriate and that was used in the final evaluation.

4.1.7 Influence of the inversion scheme

Different inversion schemes based on different models can and do produce different flux estimates. Although the aim of the IMPLICIT is not to compare multiple inversion schemes but to focus on FLEXINVERT, we have previously presented the effects of a different inversion scheme with regard to the statistical scores and the modelled time series. The fluxes change accordingly. This inversion scheme shows, as expected, no negative fluxes (Figure 4.17). As in the FLEXINVERT inversions, there is an increase in the fluxes in central UK, Ireland and the Netherlands with respect to the a priori data. This scheme produces even higher total yearly emissions for Ireland (1013 kt) than those obtained with FLEXINVERT. The time series at the Irish sites show a better agreement for the maxima compared with FLEXINVERT.

4.2 Discussion on the Sensitivity Tests

A large number of sensitivity tests have been undertaken in order to identify the most appropriate set-up for the inversion, considering the data available and associated uncertainties. Computational constraints, limiting the dimensions of the matrices and, by implication, the resolution of the runs, have also been considered. In conclusion:

- Given the relatively small size of the Irish domain, it is advisable to use a higher spatial resolution output for the SRS. However, this comes at the expense of requiring better constraining data (observations or a priori).
- Sensitivity tests on uncertainties indicate that the inversion better constrains the emissions when realistic and appropriate assumptions around the errors are used. It is well known that the atmospheric transport modelling incorporates a large contribution to the estimated flux uncertainty and this needs to be appropriately quantified.
- The two different inversion schemes demonstrate quite different behaviour. FLEXINVERT often underestimates the amplitude of the peaks whereas the log-normal approach, LOGINVERT, sometimes overestimates them, but overall it matches the observations more closely. This shows that FLEXINVERT may require additional fine-tuning of the parameters used to define error and uncertainty characteristics.
- Using a time window for the observations instead of daily averages did not significantly change emission estimates for CH_4. For other species with strong diurnal cycles, this may not be the case.
- For the Irish domain, performing the inversion with all of the stations or with the subset_sites makes little difference, with emissions of 952 kt and 940 kt, respectively (less than a 1.5% difference), with a similar geographical distribution. When we consider only the UK and Irish sites in the inversions, we see that the flux is slightly modified (by about 2.6%), whereas, when Carnsore Point is not used, the overall flux in Ireland decreases (by about 6%).
- A correlation length of 100 km appears to be sufficient. A much smaller correlation length of 10 km has been tested without large differences in how the outcome represents the time series at the observational sites.

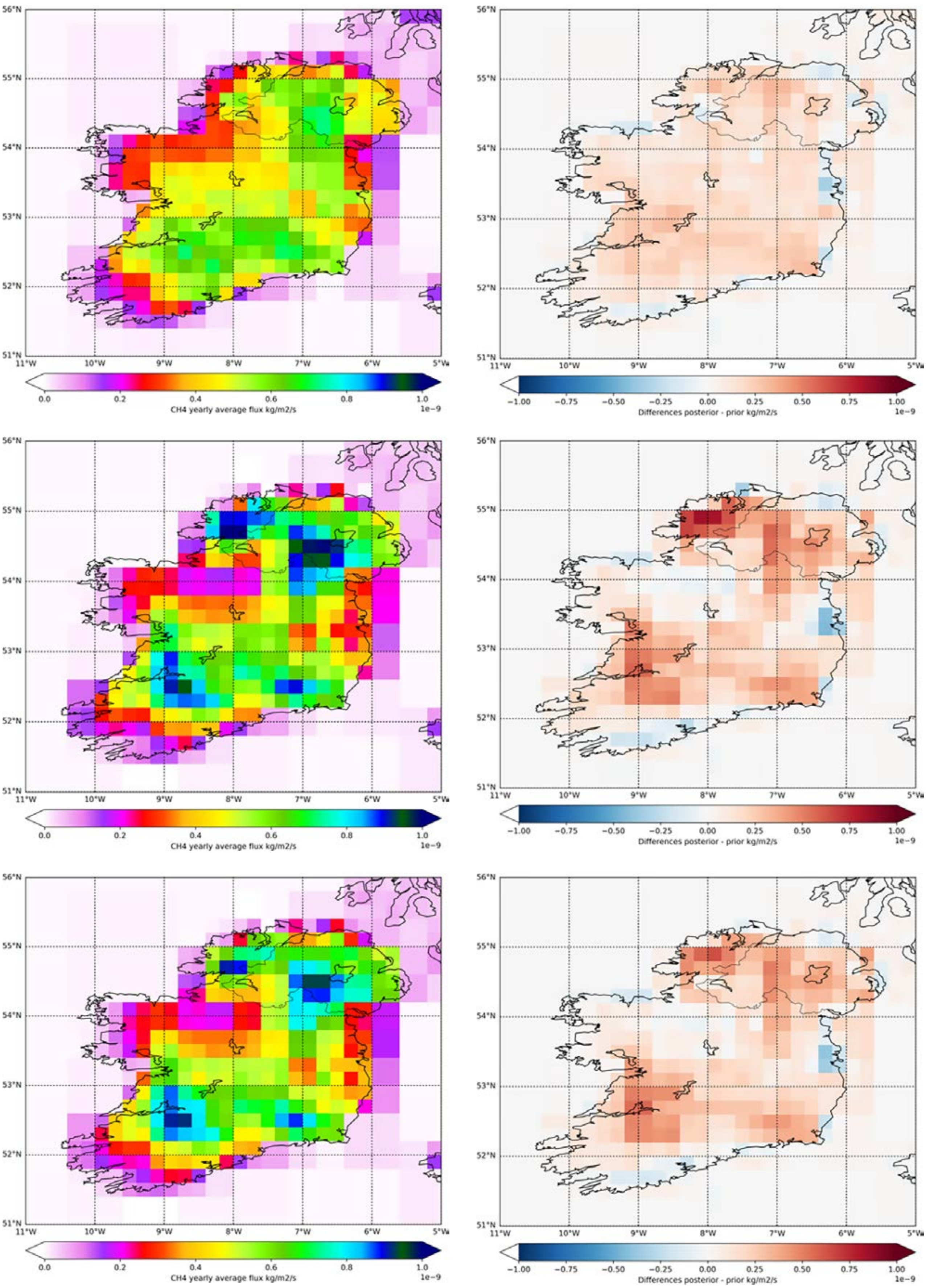

Figure 4.11. Posterior average flux (left) and differences with respect to the a priori (right) for the 0.2 degree SRS files (SRS0.2×0.2) using the 0.1 degree EDGAR emission inventory (EDGAR_0.1×0.1_ANT) for the basic test (upper panels), test 1 (middle panels) and test 2 (bottom panels).

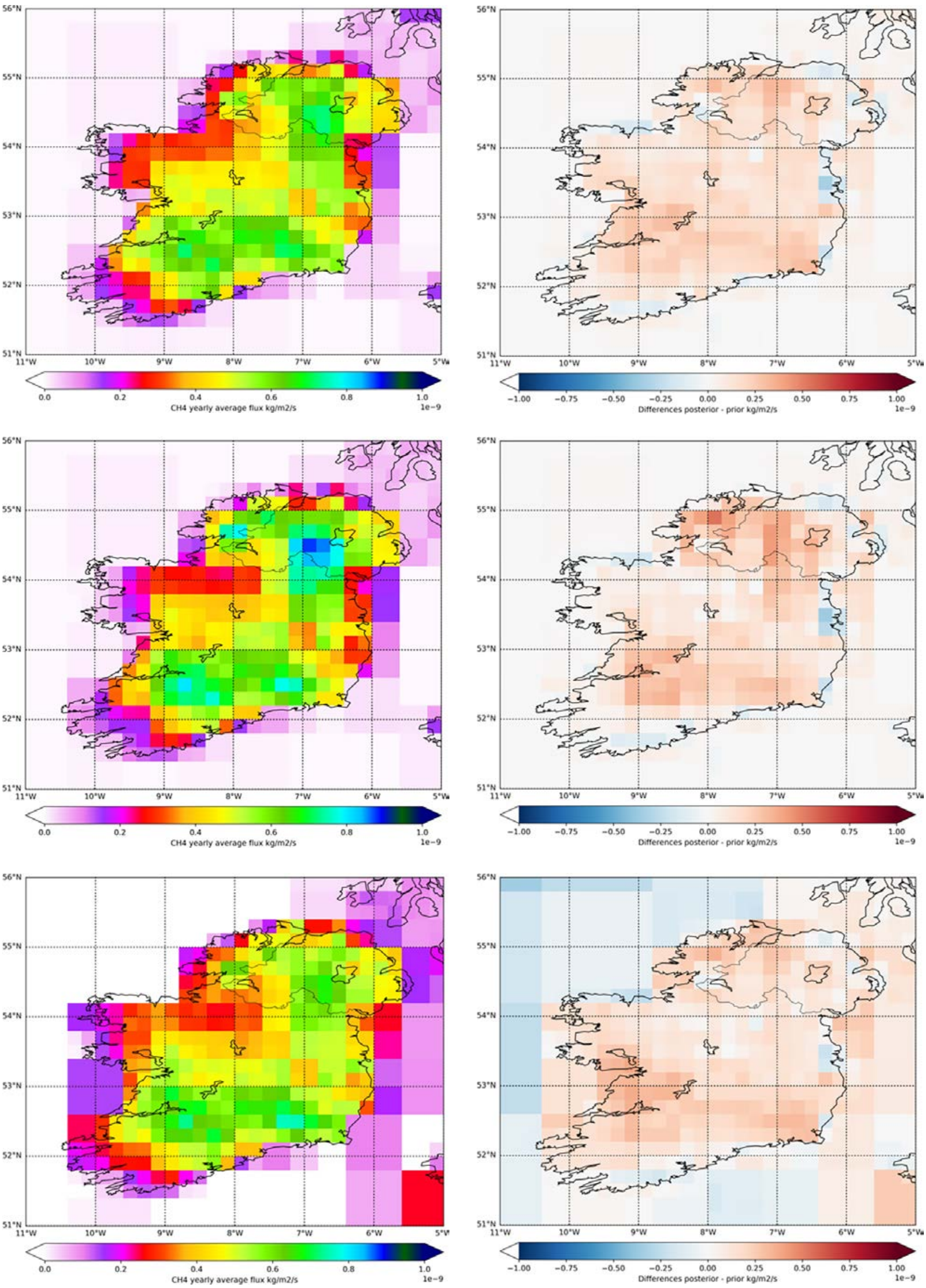

Figure 4.12. Posterior average flux (left) and differences with respect to the a priori (right) for the 0.2 degree SRS files (SRS0.2×0.2) using the 0.1 degree EDGAR emission inventory (EDGAR_0.1×0.1_ANT) for the basic test (upper panels), test 3 (middle panels) and test 4 (bottom panels).

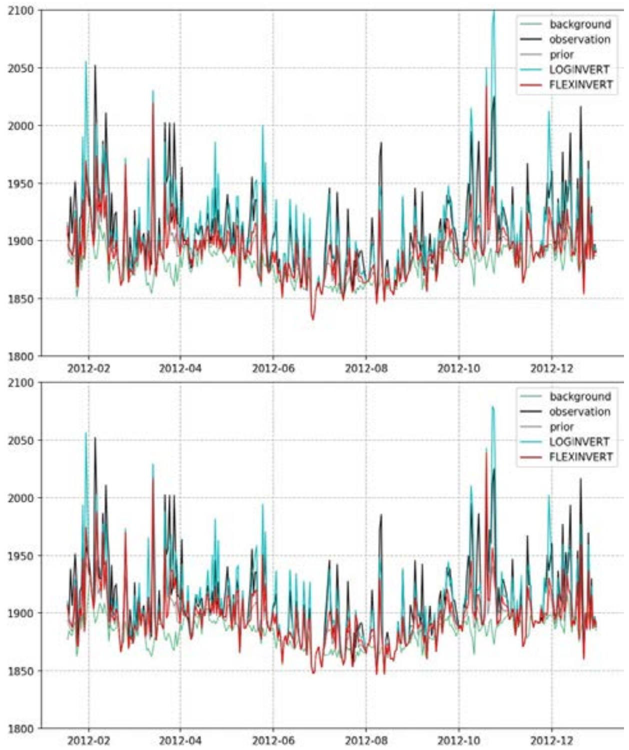

Figure 4.13. Time series for the basic test (upper panel) and test 4 (lower panel) for Carnsore Point.

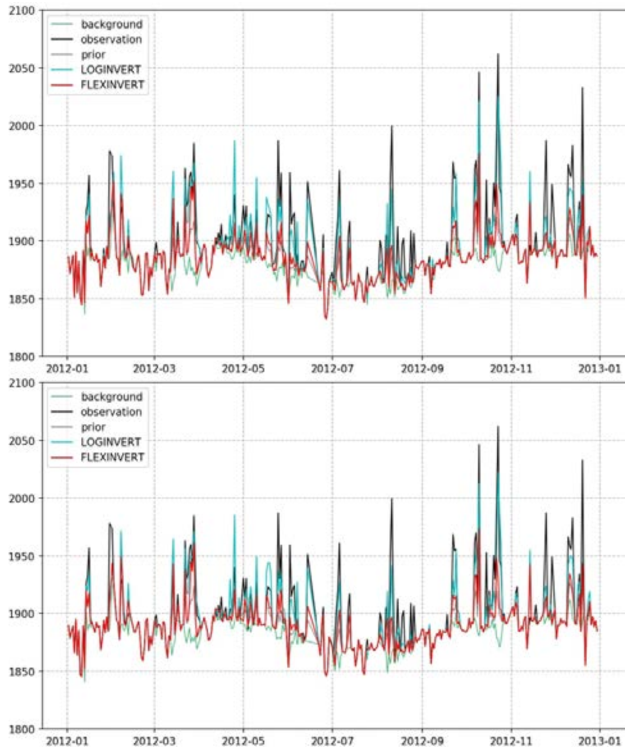

Figure 4.14. Time series for the basic test (upper panel) and test 4 (upper panel) for Mace Head.

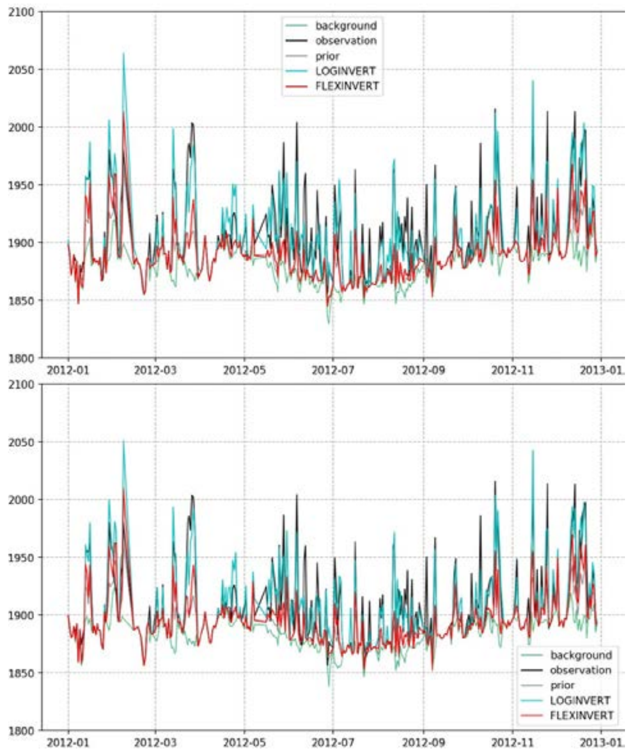

Figure 4.15. Time series for the basic test (upper panel) and test 4 (lower panel) for Malin Head.

Table 4.2. Statistical scores and their averages for all of the sites used in the inversion for the basic test and tests 1–4

Site	A priori			FLEXINVERT			LOGINVERT		
	NSD	*R*	RMSE	NSD	*R*	RMSE	NSD	*R*	RMSE
Basic test									
Malin_h	0.53	0.71	30.82	0.66	0.76	27.41	1.01	0.89	15.92
Sonnbli	0.98	0.70	17.22	0.98	0.72	16.72	1.24	0.79	13.20
Neuglob	0.53	0.65	56.65	0.70	0.68	45.15	1.19	0.76	33.86
Jungfra	0.78	0.83	14.03	0.79	0.82	15.08	1.02	0.85	13.23
Carnsor	0.54	0.81	30.32	0.69	0.83	25.55	1.05	0.84	21.07
Puy_de_	0.81	0.63	20.53	0.78	0.70	22.02	1.10	0.79	17.08
Schauin	0.72	0.65	26.61	0.83	0.60	27.08	1.10	0.70	22.33
Zugspit	0.68	0.78	22.29	0.72	0.77	22.68	0.87	0.76	18.71
Plateau	0.94	0.92	8.007	0.94	0.89	9.615	1.21	0.88	12.35
Hohenpe	0.43	0.57	57.56	0.65	0.80	31.39	0.63	0.68	35.50
Ridge_H	0.40	0.54	54.88	0.60	0.61	45.89	0.94	0.65	40.13
Monte_C	0.54	0.35	28.68	0.55	0.34	28.92	0.96	0.66	20.21
Mace_he	0.52	0.75	25.91	0.62	0.82	21.95	0.86	0.88	15.55
Average	0.66	0.68	30.27	0.73	0.72	26.11	1.10	0.78	21.47
Test 1									
Malin_h	0.53	0.71	31.04	0.67	0.76	27.37	1	0.89	16.04
Sonnbli	0.98	0.70	17.15	1.00	0.72	16.86	1.24	0.79	13.28
Neuglob	0.53	0.65	56.70	0.73	0.69	45.32	1.20	0.76	33.85
Jungfra	0.78	0.83	14.02	0.81	0.83	14.89	1.02	0.85	13.29
Carnsor	0.55	0.81	30.53	0.72	0.83	25.12	1.04	0.84	20.87
Puy_de_	0.81	0.62	20.63	0.78	0.70	22.52	1.11	0.79	17.45
Schauin	0.73	0.65	26.56	0.86	0.60	26.92	1.12	0.70	22.58
Zugspit	0.69	0.78	22.31	0.74	0.77	23.03	0.87	0.77	18.56
Plateau	0.93	0.92	7.975	0.96	0.88	10.43	1.18	0.89	11.41
Hohenpe	0.44	0.57	57.84	0.62	0.69	36.39	0.59	0.68	37.23
Ridge_H	0.40	0.53	55.34	0.53	0.60	47.31	0.94	0.65	40.38
Monte_C	0.54	0.35	28.67	0.58	0.32	29.15	0.96	0.65	20.36
Mace_he	0.51	0.75	26.17	0.61	0.83	21.98	0.85	0.88	15.83
Average	0.65	0.68	30.38	0.74	0.71	26.72	1.01	0.78	21.63
Test 2									
Malin_h	0.51	0.72	30.31	0.66	0.77	26.45	0.99	0.90	15.08
Sonnbli	0.97	0.70	16.53	0.98	0.72	16.16	1.23	0.78	13.19
Neuglob	0.53	0.67	55.53	0.70	0.70	45.48	1.10	0.79	30.03
Jungfra	0.78	0.83	13.78	0.81	0.83	14.42	1.02	0.84	13.45
Carnsor	0.54	0.82	29.60	0.71	0.84	24.23	1.03	0.84	20.33
Puy_de_	0.79	0.65	19.74	0.76	0.72	21.64	1.10	0.81	16.42
Schauin	0.73	0.65	25.89	0.84	0.62	26.10	1.13	0.68	23.36
Zugspit	0.68	0.79	21.81	0.73	0.77	22.33	0.88	0.77	18.45
Plateau	0.94	0.92	7.842	0.95	0.89	9.498	1.19	0.88	11.61
Hohenpe	0.42	0.55	57.29	0.58	0.75	35.99	0.57	0.68	36.96
Ridge_H	0.40	0.53	54.52	0.55	0.62	45.78	0.92	0.65	39.8
Monte_C	0.54	0.37	27.89	0.56	0.37	27.98	0.96	0.67	19.56
Mace_he	0.50	0.75	25.70	0.61	0.83	21.54	0.84	0.88	15.54
Average	0.64	0.69	29.73	0.73	0.73	25.97	1.00	0.78	21.06

Table 4.2. Continued

Site	A priori			FLEXINVERT			LOGINVERT		
	NSD	*R*	RMSE	NSD	*R*	RMSE	NSD	*R*	RMSE
Test 3									
Malin_h	0.53	0.71	30.74	0.65	0.75	27.63	1.08	0.87	18.10
Sonnbli	0.99	0.70	17.59	1.00	0.71	17.15	1.24	0.79	13.24
Neuglob	0.53	0.65	56.52	0.67	0.69	47.57	1.14	0.78	32.00
Jungfra	0.79	0.84	14.05	0.81	0.83	14.91	1.01	0.85	13.17
Carnsor	0.54	0.81	30.33	0.68	0.83	25.77	1.17	0.81	25.06
Puy_de_	0.81	0.63	20.58	0.78	0.69	22.26	1.14	0.79	17.56
Schauin	0.73	0.65	26.74	0.85	0.61	27.19	1.11	0.70	22.59
Zugspit	0.69	0.79	22.49	0.73	0.77	22.94	0.88	0.77	18.72
Plateau	0.93	0.92	7.992	0.95	0.89	9.731	1.20	0.88	12.44
Hohenpe	0.43	0.56	57.83	0.61	0.78	34.32	0.60	0.68	36.37
Ridge_H	0.40	0.53	54.90	0.54	0.61	47.12	1.04	0.61	43.97
Monte_C	0.55	0.36	28.80	0.57	0.33	29.25	0.96	0.67	20.00
Mace_he	0.52	0.75	25.91	0.59	0.82	22.67	0.91	0.88	15.48
Average	0.65	0.68	30.34	0.73	0.72	26.81	1.04	0.77	22.21
Test 4									
Malin_h	0.51	0.73	28.49	0.66	0.79	23.76	1.00	0.89	15.09
Sonnbli	0.98	0.72	15.37	0.97	0.74	15.13	1.23	0.78	13.34
Neuglob	0.52	0.67	53.18	0.64	0.70	44.16	1.07	0.79	28.78
Jungfra	0.79	0.84	13.15	0.80	0.83	13.98	1.04	0.85	13.27
Carnsor	0.54	0.82	27.61	0.70	0.84	22.91	1.03	0.85	19.70
Puy_de_	0.79	0.66	19.08	0.76	0.72	21.02	1.08	0.82	16.07
Schauin	0.73	0.66	24.43	0.79	0.64	25.14	1.11	0.70	22.45
Zugspit	0.70	0.80	20.37	0.73	0.79	20.96	0.91	0.78	17.78
Plateau	0.93	0.95	6.451	0.94	0.91	8.471	1.14	0.94	8.349
Hohenpe	0.42	0.52	55.66	0.59	0.79	34.00	0.57	0.66	35.97
Ridge_H	0.39	0.54	52.47	0.57	0.60	44.81	0.89	0.66	38.90
Monte_C	0.55	0.38	26.94	0.56	0.36	27.31	0.96	0.68	18.91
Mace_he	0.50	0.75	24.41	0.61	0.83	20.36	0.84	0.88	15.19
Average	0.64	0.70	28.28	0.72	0.73	24.77	0.99	0.79	20.29

4.3 Final Estimates

From the results of the sensitivity tests, we sought to further investigate the inversion estimates from the test 4 set-up, using the 0.2 degree SRS files (SRS0.2×0.2) and two a priori emissions inventories.

4.3.1 "All sites" and "all sites with half of the Irish data"

The inversion results using all of the available data and using all sites with half of the Irish data yielded quite similar emissions (Table 4.3) and spatial distributions (Figure 4.18).

The statistical scores for the independent set of data from the Irish sites (Tables 4.4 and 4.5) show that the increase in the correlation is very small when using the source term estimated by FLEXINVERT. The RMSE is also not modified very much, but there is a larger decrease when the EDGAR_0.1×0.1_ANT emission inventory is used. The more flexible LOGINVERT system has an improved correlation, decreased RMSE and increased NSD. It is clear that in the current set-up, and with the data available, FLEXINVERT is largely influenced by the a priori information. The statistics for all of the remaining sites and data show a similar behaviour, with a larger increase in the correlation when using the high-resolution a priori

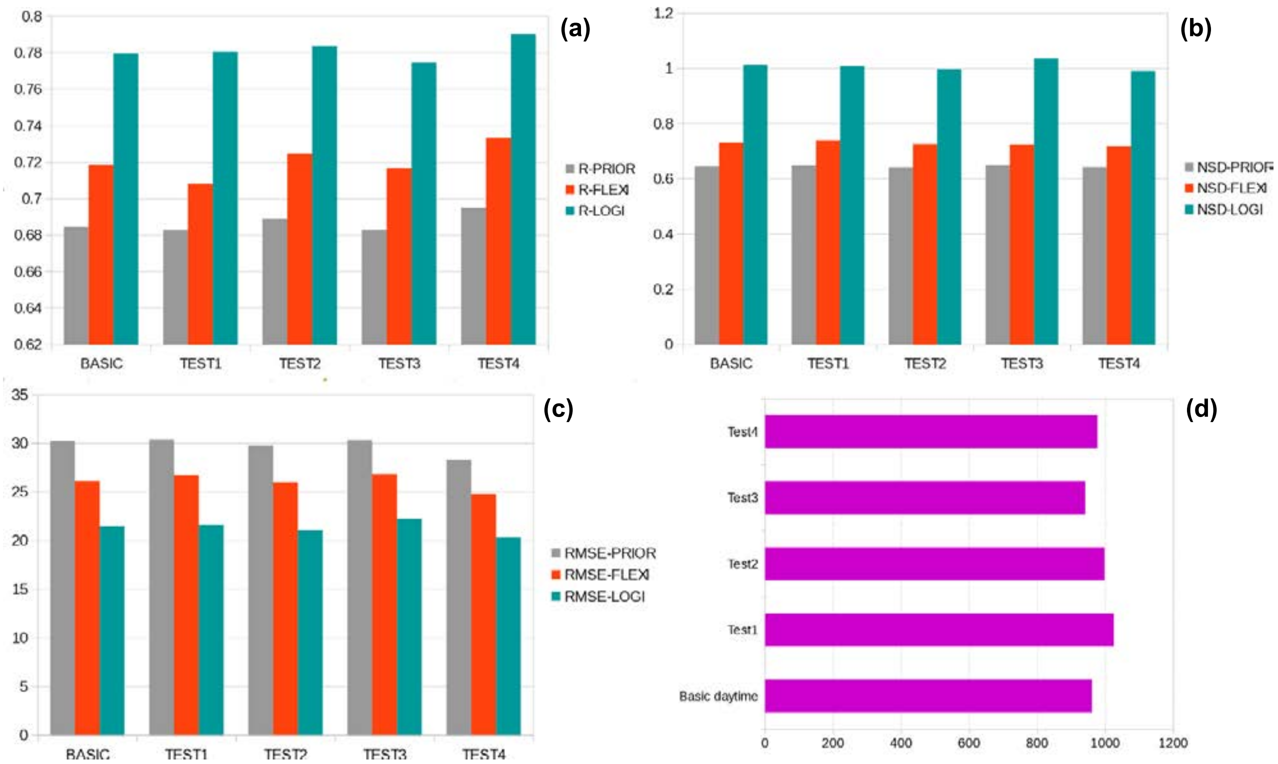

Figure 4.16. Average scores for the five tests at the three Irish sites and total emissions obtained. (a) *R*; (b) NSD; (c) RMSE; and (d) total emissions (kt).

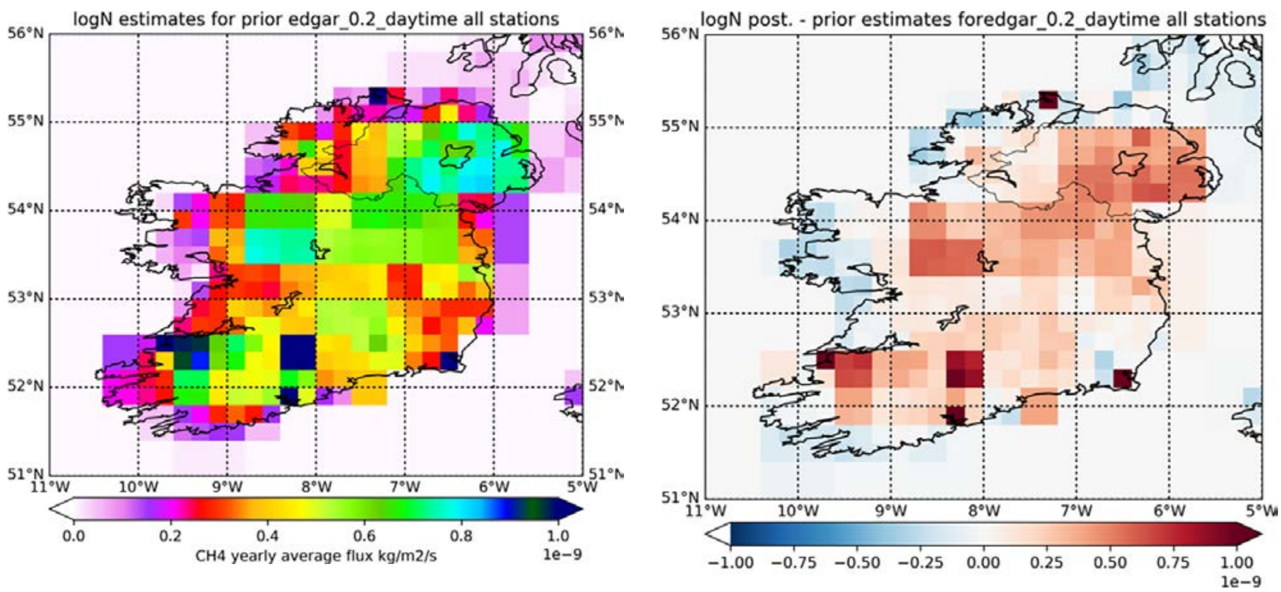

Figure 4.17. Posterior yearly average flux (left) and differences with respect to the a priori (right) (EDGAR_0.1×0.1_ANT) for LOGINVERT.

Table 4.3. Total emissions for each of the inversions using two inventories and the “all_sites” and “all_ sites –half of the Irish data” runs

Inventory	Total 2012 emissions (kt)	
	All sites	All sites – half of the Irish data
EDGAR_1.0×1.0_NAT	938.4	872.2
EDGAR_0.1×0.1_ANT	963.5	895.8

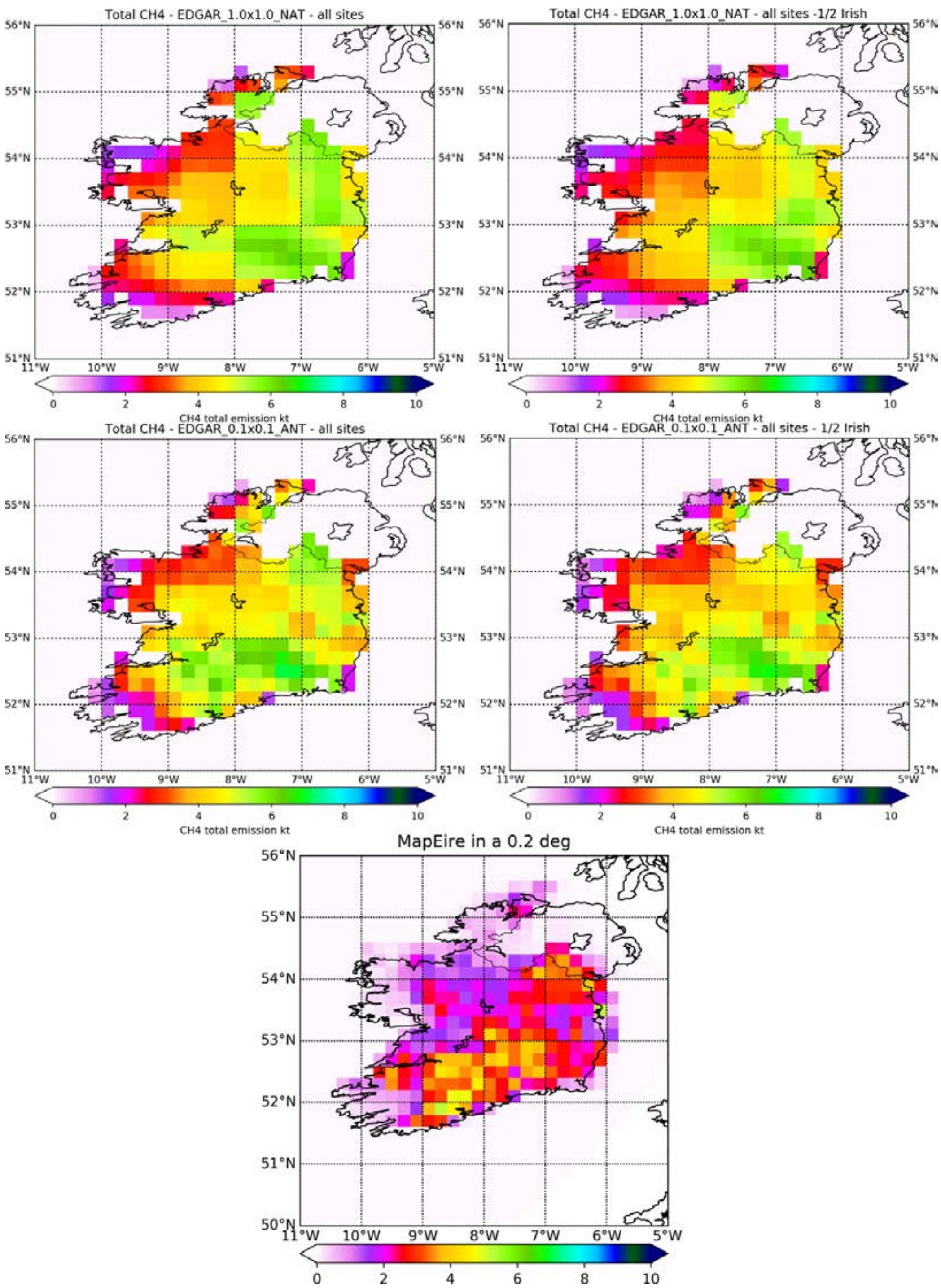

Figure 4.18. Total emission maps for the different runs using the two emissions inventories as a priori and using all of the sites (left panels) or all of the sites with half of the Irish data (right panels) compared with the MapEIre emission inventory for 2015 (bottom panel).

Table 4.4. Statistical scores and their averages for the Irish sites (using only the data not used in the inversion) and the EDGAR_1.0×1.0_ANT inventory

Site	A priori			FLEXINVERT			LOGINVERT		
	NSD	*R*	RMSE	NSD	*R*	RMSE	NSD	*R*	RMSE
Malin_h	0.52	0.70	29.37	0.60	0.73	27.17	0.84	0.88	16.12
Carnsor	0.56	0.78	25.88	0.68	0.81	21.81	0.95	0.81	19.27
Mace_he	0.50	0.81	25.31	0.57	0.84	22.91	0.74	0.86	18.57
Average	0.53	0.76	26.85	0.62	0.79	26.96	0.84	0.85	17.99

Table 4.5. Statistical scores and their averages for the Irish sites (using only the data not used in the inversion) and the EDGAR_0.1×0.1_NAT inventory

Site	A priori			FLEXINVERT			LOGINVERT		
	NSD	*R*	RMSE	NSD	*R*	RMSE	NSD	*R*	RMSE
Malin_h	0.51	0.70	29.39	0.60	0.74	26.79	0.89	0.87	16.49
Carnsor	0.54	0.79	25.66	0.68	0.81	27.73	1.06	0.79	20.96
Mace_he	0.49	0.81	25.27	0.58	0.86	22.25	0.85	0.90	15.15
Average	0.52	0.77	26.77	0.62	0.80	25.59	0.93	0.85	17.53

EDGAR_0.1×0.1_NAT emission inventory (Tables 4.6 and 4.7). A visual representation of the averaged statistical scores is provided in Figure 4.19. It is also clear there that, although there is an improvement in the scores, it is not significant.

The time series of the runs with all of the sites (Figure 4.20) support the scores obtained in Tables 4.4–4.7. The time series of the modelled concentrations using the FLEXINVERT estimates do show some differences for some of the dates; however, they tend to follow the a priori. In contrast, the LOGINVERT system seems to sometimes capture a better representation of the maximum concentration of CH_4 in the observational sites at the expense, however, of increasing even more the total emissions for the Irish domain.

4.3.2 "ie_uk_sites"

These simulations allow for comparison with a larger set of independent data. In this case, the overall total emissions are higher than reported anthropogenic national emissions and those obtained in the MapEIre project for the 2015 emissions. The geographical distribution of the estimated emissions, focusing on the EDGAR_0.1×0.1_NAT runs, is very similar to that seen for "all sites", but with slightly lower emissions in the area to the south of Northern Ireland and higher emissions to the north-east of Galway and north of Dublin. Table 4.8 shows the total 2012 emissions using the "ie_uk_sites" for the two emission inventories and Figure 4.21 illustrates these emissions compared with the MapEIre emission inventory.

As in the previous evaluation, the statistical scores (Tables 4.9–4.11) show a slight improvement in the results for the estimated flux with FLEXINVERT, but this does not exceed a 5% improvement. The LOGINVERT system again shows a better agreement with the observations. The time series at the three Irish observation sites for this set of simulations is shown in Figure 4.22.

Table 4.6. Statistical scores and their averages for all of the sites and data used in the inversion and the two inventories

Site	A priori			FLEXINVERT			LOGINVERT		
	NSD	*R*	RMSE	NSD	*R*	RMSE	NSD	*R*	RMSE
EDGAR_1.0. ×1.0_NAT									
Malin_h	0.53	0.75	27.66	0.65	0.80	23.95	0.92	0.91	14.30
Sonnbli	1.03	0.70	15.47	0.98	0.73	15.21	1.28	0.76	14.57
Neuglob	0.52	0.66	51.57	0.60	0.71	46.23	1.01	0.80	27.99
Jungfra	0.81	0.83	13.23	0.80	0.82	14.28	1.03	0.84	13.39
Carnsor	0.55	0.82	28.96	0.69	0.84	24.74	0.96	0.88	18.20
Puy_de_	0.84	0.68	18.61	0.74	0.70	20.49	1.10	0.78	17.94
Schauin	0.79	0.63	23.50	0.80	0.66	25.16	1.04	0.72	20.87
Zugspit	0.72	0.80	19.70	0.74	0.79	20.65	0.91	0.79	17.41
Plateau	0.95	0.94	6.75	0.96	0.89	9.49	1.16	0.93	9.18
Hohenpe	0.44	0.61	49.27	0.62	0.81	31.50	0.53	0.72	35.37
Ridge_H	0.46	0.57	48.83	0.59	0.59	45.09	0.85	0.67	38.24
Monte_C	0.55	0.38	26.78	0.56	0.43	26.29	0.95	0.69	18.69
Mace_he	0.52	0.70	23.33	0.57	0.76	21.47	0.73	0.87	15.64
Average	0.67	0.70	27.20	0.72	0.73	24.97	0.96	0.80	20.14
EDGAR_0.1×0.1_ANT									
Malin_h	0.51	0.75	27.92	0.66	0.81	23.40	1.03	0.90	14.88
Sonnbli	0.98	0.72	15.37	0.98	0.73	15.08	1.26	0.76	14.28
Neuglob	0.52	0.67	53.18	0.59	0.70	46.29	1.01	0.80	27.50
Jungfra	0.79	0.84	13.15	0.81	0.82	14.28	1.06	0.83	14.21
Carnsor	0.54	0.84	28.74	0.69	0.85	24.32	1.09	0.86	20.63
Puy_de_	0.79	0.66	19.08	0.75	0.71	19.10	1.08	0.81	16.81
Schauin	0.73	0.66	24.43	0.80	0.65	25.22	1.17	0.65	25.26
Zugspit	0.70	0.80	20.37	0.74	0.79	20.58	0.95	0.75	18.67
Plateau	0.93	0.95	6.45	0.96	0.89	9.43	1.15	0.94	8.50
Hohenpe	0.42	0.52	55.66	0.61	0.82	31.15	0.59	0.62	35.60
Ridge_H	0.39	0.54	52.47	0.55	0.58	45.90	0.83	0.65	39.15
Monte_C	0.55	0.38	26.94	0.56	0.40	26.66	0.99	0.69	19.16
Mace_he	0.51	0.69	23.67	0.58	0.77	21.11	0.86	0.88	14.26
Average	0.52	0.76	26.78	0.64	0.81	22.94	0.99	0.88	16.59

Table 4.7. Statistical scores and their averages using the “all_sites” runs and the two inventories

Site	A priori			FLEXINVERT			LOGINVERT		
	NSD	*R*	RMSE	NSD	*R*	RMSE	NSD	*R*	RMSE
EDGAR_1.0×1.0_NAT									
Malin_h	0.49	0.76	32.10	0.61	0.81	27.12	0.95	0.90	15.25
Sonnbli	1.09	0.74	13.42	1.08	0.76	14.72	1.44	0.78	17.14
Neuglob	0.59	0.69	48.52	0.65	0.74	40.97	1.08	0.81	24.57
Jungfra	0.86	0.85	11.21	0.91	0.86	13.30	1.16	0.86	14.24
Puy_de_	0.91	0.71	17.69	0.86	0.71	21.28	1.18	0.81	14.85
Carnsor	0.46	0.72	39.14	0.57	0.78	33.08	0.97	0.81	22.95
Ochsenk	0.86	0.73	27.80	0.92	0.74	26.13	1.94	0.75	35.49
Schauin	0.83	0.58	23.76	0.88	0.65	24.81	1.10	0.70	21.45
Zugspit	0.72	0.79	20.90	0.76	0.80	21.58	0.98	0.73	18.66

Table 4.7. Continued

Site	A priori			FLEXINVERT			LOGINVERT		
	NSD	*R*	RMSE	NSD	*R*	RMSE	NSD	*R*	RMSE
Plateau	1.10	0.90	9.63	1.06	0.90	9.25	1.47	0.81	17.95
Hohenpe	0.48	0.66	48.09	0.65	0.83	31.38	0.57	0.73	34.06
Ridge_H	0.60	0.71	37.67	0.75	0.74	31.62	1.11	0.82	23.01
Monte_C	0.60	0.41	27.17	0.62	0.50	26.14	1.02	0.72	17.05
Mace_he	0.54	0.76	23.62	0.61	0.83	20.36	0.90	0.89	13.44
Average	0.72	0.71	27.19	0.78	0.76	24.41	1.13	0.79	20.72
EDGAR_0.1×0.1_ANT									
Malin_h	0.47	0.76	32.46	0.60	0.82	26.91	0.95	0.91	15.23
Sonnbli	1.03	0.76	14.46	1.07	0.76	14.52	1.28	0.83	12.29
Neuglob	0.58	0.70	51.03	0.65	0.74	40.96	1.06	0.82	23.67
Jungfra	0.84	0.88	10.76	0.89	0.87	12.43	1.08	0.89	10.93
Puy_de_	0.86	0.69	18.08	0.85	0.72	19.59	1.10	0.84	12.91
Carnsor	0.45	0.74	38.47	0.56	0.78	33.31	0.91	0.81	23.46
Ochsenk	0.87	0.74	28.21	0.86	0.73	26.09	1.57	0.78	26.29
Schauin	0.76	0.64	24.55	0.87	0.65	24.47	1.14	0.67	23.16
Zugspit	0.71	0.80	22.34	0.76	0.80	21.17	0.92	0.75	18.09
Plateau	1.00	0.97	4.89	1.02	0.93	7.33	1.22	0.91	10.00
Hohenpe	0.45	0.56	55.94	0.64	0.84	30.77	0.59	0.62	36.37
Ridge_H	0.51	0.68	43.16	0.71	0.74	32.62	0.99	0.82	23.52
Monte_C	0.60	0.38	28.05	0.62	0.42	27.35	0.93	0.72	16.78
Mace_he	0.51	0.75	24.27	0.60	0.84	20.28	0.86	0.91	12.90
Average	0.69	0.72	28.33	0.76	0.76	24.13	1.04	0.80	18.97

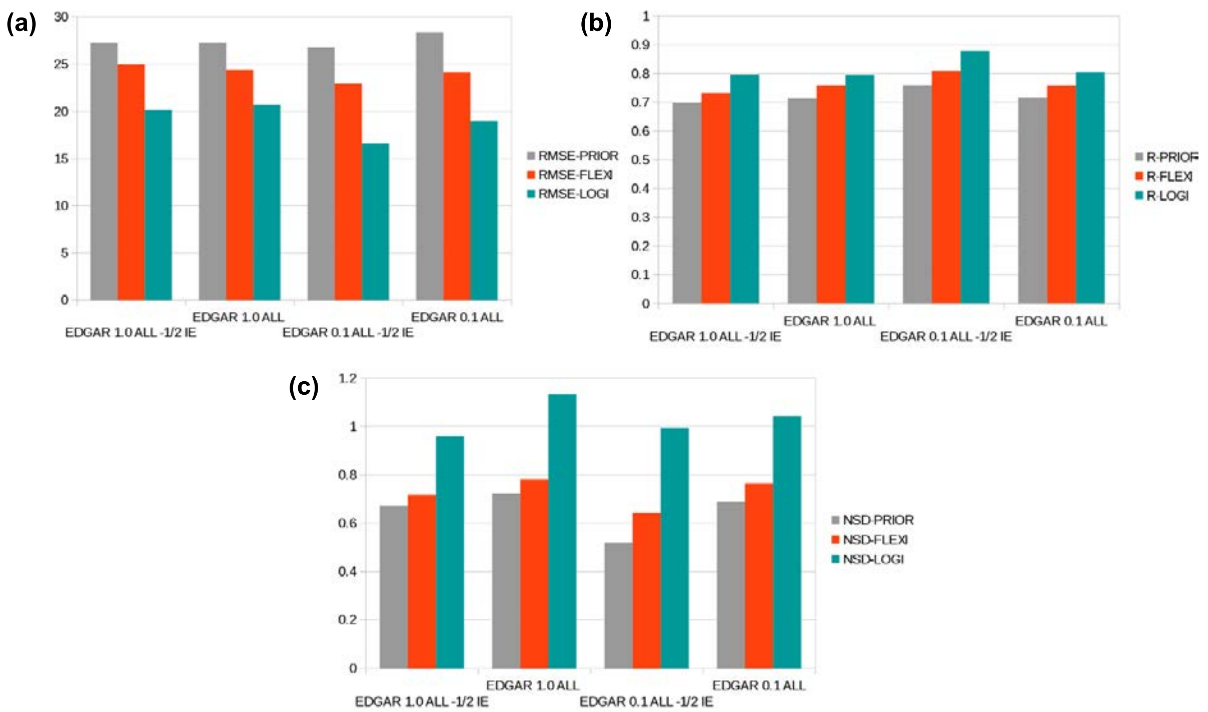

Figure 4.19. Summary of the averaged statistics for the runs including all of the sites and the runs without half of the Irish data for the dependent sites (i.e. sites and data used in the actual inversions). (a) RMSE; (b) *R*; and (c) NSD.

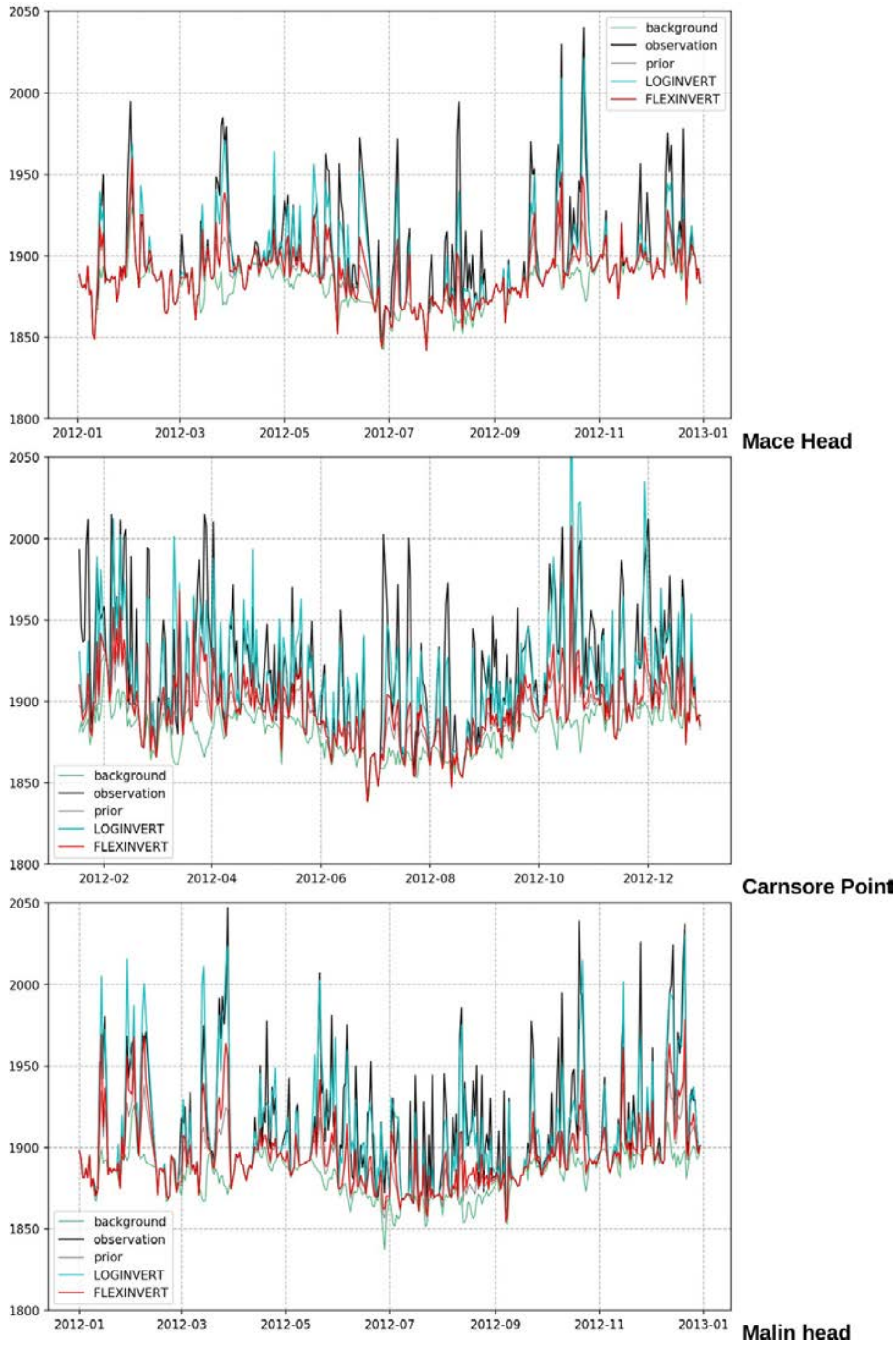

Figure 4.20. Time series at Mace Head (top), Carnsore Point (middle) and Malin Head (bottom) with the observed values (black), values modelled using the a priori as the emission inventory (grey), the FLEXINVERT estimates (red) and the LOGINVERT estimates (blue).

Table 4.8. Total emissions for each of the inversions using two inventories and the “ie_uk_sites” runs

Inventory	Total 2012 emissions (kt)
	Irish + UK sites
EDGAR_1.0×1.0_NAT	947.7
EDGAR_0.1×0.1_ANT	971.9

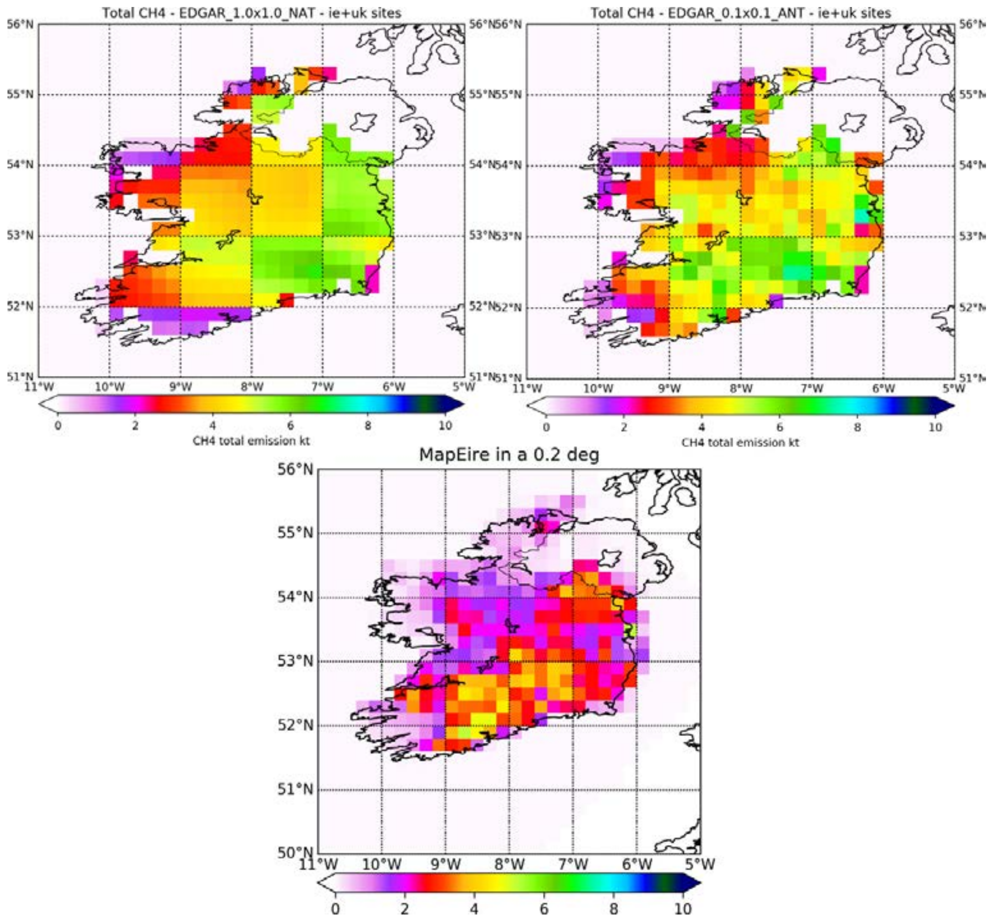

Figure 4.21. Total emission maps for the "ie_uk_sites" run using the EDGAR_1.0×1.0_NAT emission inventory (left) and the EDGAR_0.1×0.1_ANT emission inventory (middle) as a priori compared with the MapEIre emission inventory for 2015 (right).

Table 4.9. Statistical scores and their averages for the "ie_uk_sites" and the EDGAR_1.0×1.0_NAT inventory

Site	A priori			FLEXINVERT			LOGINVERT		
	NSD	*R*	RMSE	NSD	*R*	RMSE	NSD	*R*	RMSE
Malin_h	0.54	0.80	29.14	0.62	0.82	26.29	0.98	0.91	14.87
Carnsor	0.49	0.74	37.54	0.58	0.79	32.43	1.00	0.81	23.65
Ridge_H	0.64	0.75	36.50	0.77	0.75	30.51	1.13	0.83	22.66
Mace_he	0.54	0.77	23.43	0.62	0.84	20.05	0.93	0.87	14.95
Average	0.55	0.76	31.65	0.65	0.80	27.32	1.01	0.85	19.03

Table 4.10. Statistical scores and their averages for the "ie_uk_sites" and the EDGAR_0.1×0.1_ANT inventory

Site	A priori			FLEXINVERT			LOGINVERT		
	NSD	*R*	RMSE	NSD	*R*	RMSE	NSD	*R*	RMSE
Malin_h	0.47	0.74	32.97	0.62	0.83	26.09	0.98	0.92	14.24
Carnsor	0.47	0.72	38.67	0.57	0.78	32.84	0.96	0.81	23.12
Ridge_H	0.60	0.71	37.29	0.72	0.74	31.48	1.01	0.82	22.59
Mace_he	0.53	0.78	23.05	0.61	0.84	20.05	0.78	0.91	14.22
Average	0.52	0.74	33.00	0.63	0.80	27.62	0.93	0.86	18.54

Table 4.11. Statistical scores and their averages for the "ie_uk_sites" runs but against the independent data of the remaining stations and the two emissions inventories used as a priori

Site	A priori			FLEXINVERT			LOGINVERT		
	NSD	*R*	RMSE	NSD	*R*	RMSE	NSD	*R*	RMSE
EDGAR_1.0×1.0_NAT									
Sonnbli	1.06	0.76	12.94	1.10	0.76	13.08	1.43	0.76	16.66
Neuglob	0.55	0.69	49.43	0.60	0.69	47.35	0.97	0.79	26.87
Jungfra	0.84	0.89	10.52	0.90	0.86	11.07	1.17	0.85	14.61
Puy_de_	0.88	0.72	18.92	0.90	0.73	18.92	1.19	0.80	15.04
Ochsenk	0.85	0.73	27.92	0.92	0.74	27.25	1.64	0.77	28.21
Schauin	0.80	0.62	24.81	0.84	0.60	23.44	1.06	0.69	21.33
Zugspit	0.72	0.79	21.58	0.74	0.78	20.78	0.93	0.73	19.10
Plateau	1.02	0.95	6.01	1.20	0.85	12.35	1.50	0.79	18.82
Hohenpe	0.48	0.72	48.52	0.49	0.68	47.27	0.51	0.66	40.83
Monte_C	0.60	0.42	27.29	0.64	0.51	24.93	1.04	0.69	18.15
Average	0.78	0.73	24.79	0.83	0.72	24.64	1.14	0.75	21.96
EDGAR_0.1×0.1_ANT									
Sonnbli	1.02	0.76	14.70	1.05	0.77	14.04	1.21	0.85	10.09
Neuglob	0.57	0.71	46.89	0.59	0.69	46.27	1.14	0.79	28.03
Jungfra	0.84	0.88	11.70	0.86	0.88	10.49	0.97	0.90	9.26
Puy_de_	0.85	0.71	20.08	0.88	0.73	18.24	1.07	0.84	13.18
Ochsenk	0.82	0.74	27.84	0.85	0.72	27.06	1.39	0.80	22.43
Schauin	0.77	0.66	25.73	0.80	0.63	23.53	1.04	0.77	17.83
Zugspit	0.71	0.80	22.91	0.73	0.80	21.38	0.84	0.80	18.14
Plateau	0.98	0.97	4.72	1.03	0.96	5.68	1.16	0.93	8.25
Hohenpe	0.47	0.66	52.73	0.49	0.64	49.31	0.53	0.64	44.14
Monte_C	0.60	0.31	29.49	0.62	0.40	27.50	0.81	0.68	18.28
Average	0.76	0.72	25.68	0.79	0.72	24.35	1.02	0.80	18.96

4.3.3 Monthly evaluation for "all sites"

A monthly estimate of emissions was also performed. The results are shown in Figure 4.23a–l.

Overall, the same trend is noted as for the annual data, with a general correction upwards of the CH_4 flux but with a different geographical distribution depending on the month and the increase predominantly in the central and southern provinces. In January (see Figure 4.23a) there was a mild correction upwards in the areas of Shannon and Cork/Kerry and south of Leinster, whereas the central areas of the Leinster and Munster provinces had a slight negative correction with respect to the a priori, with a much larger correction in the northern tip of Ulster. The month of February (see Figure 4.23b) has the peculiarity that the inversion gives negative fluxes in a curved stripe crossing Ulster and going through the intermediate region of Leinster and Connacht. Similar negative fluxes (white areas) are found in Scotland and also in March in other regions of the UK. This may in part be an effect of the early 2012 cold wave (https://en.wikipedia.org/wiki/Early_2012_European_cold_wave). In this month the inversion shows mild increases in the fluxes for the regions of Connacht and the centre of Munster. March (see Figure 4.23c) shows a more significant increase in the fluxes in the westernmost side of Munster province and in the south-west of Ireland and still mild negative corrections in the south-east of Munster and the north-central regions of Connacht and Leinster. April (see Figure 4.23d) shows mild modifications of the fluxes, with a general increase in central Ireland and a negative correction in Munster. This positive correction in the central regions of Ireland becomes more significant in May (see Figure 4.23e), where the differences are also close to 1 $kg/m^2/s$ in the very

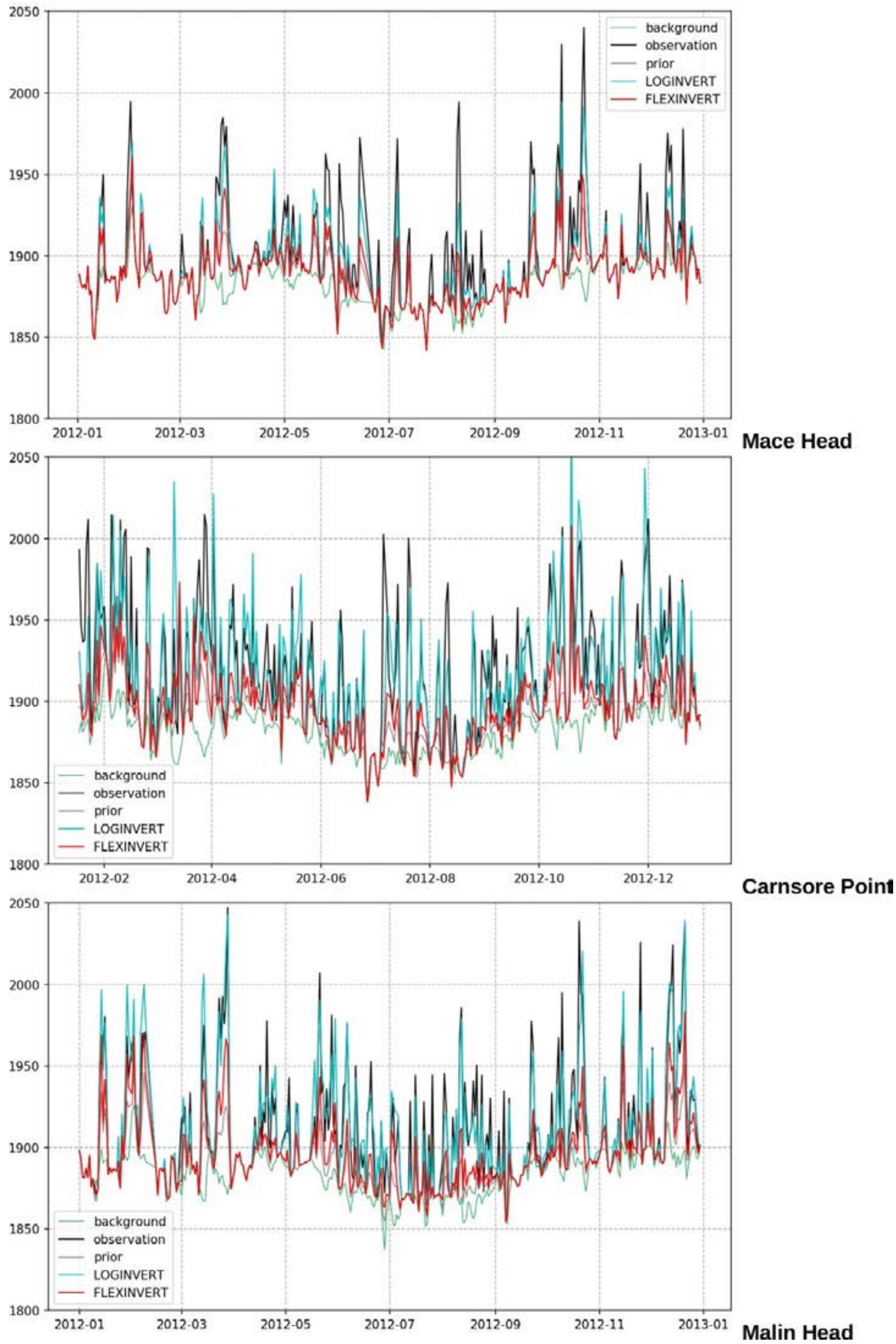

Figure 4.22. Time series at Mace Head, Carnsore Point and Malin Head with the observed values (black), values modelled using the a priori as the emission inventory (grey), the FLEXINVERT estimates (red) and the LOGINVERT estimates (blue).

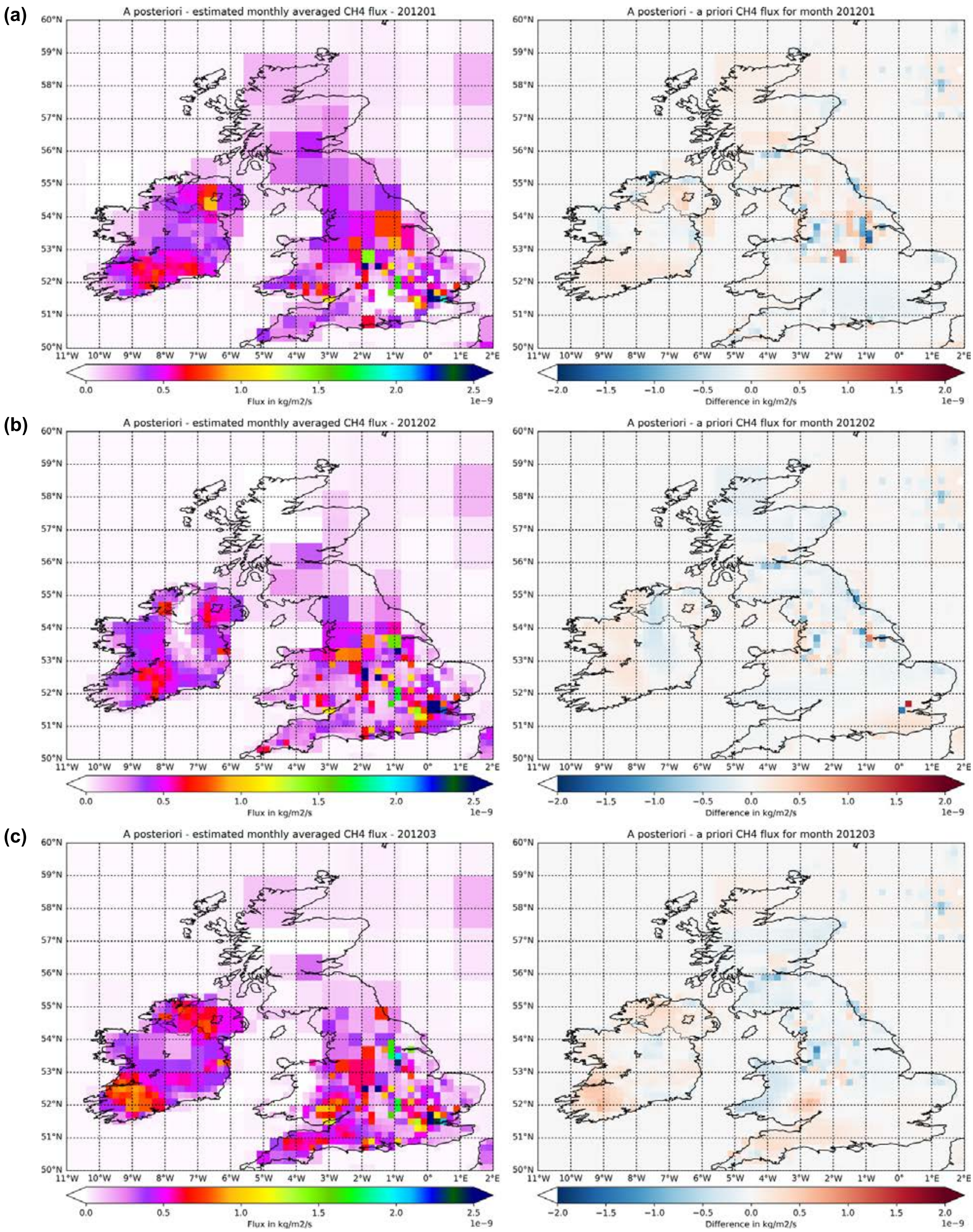

Figure 4.23. Monthly flux estimates (left) and the difference between the estimates (using the "all sites" specifications) and the a priori EDGAR_0.1×0.1_ANT (right). (a) January (b) February; (c) March.

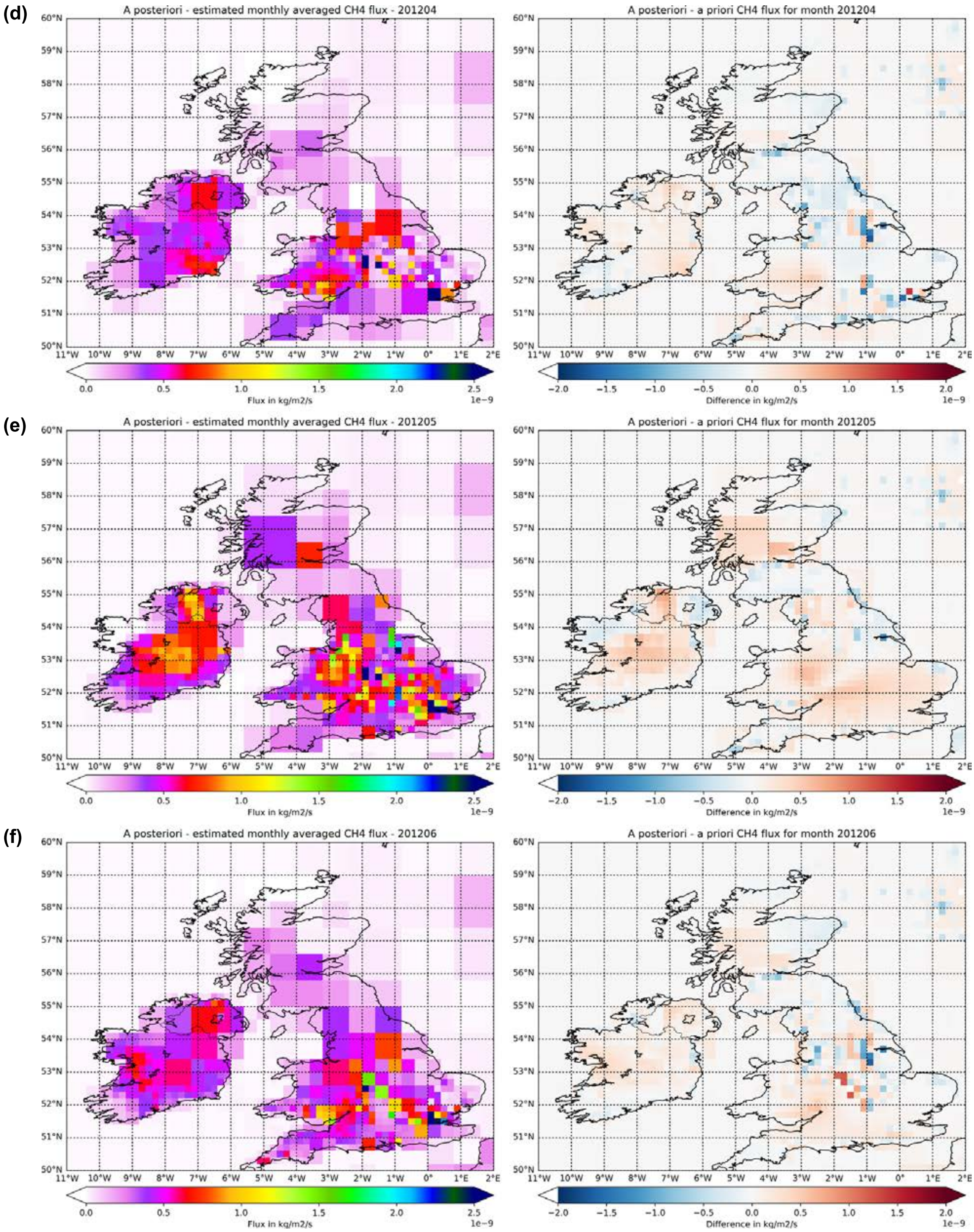

Figure 4.23. Continued. (d) April; (e) May; (f) June.

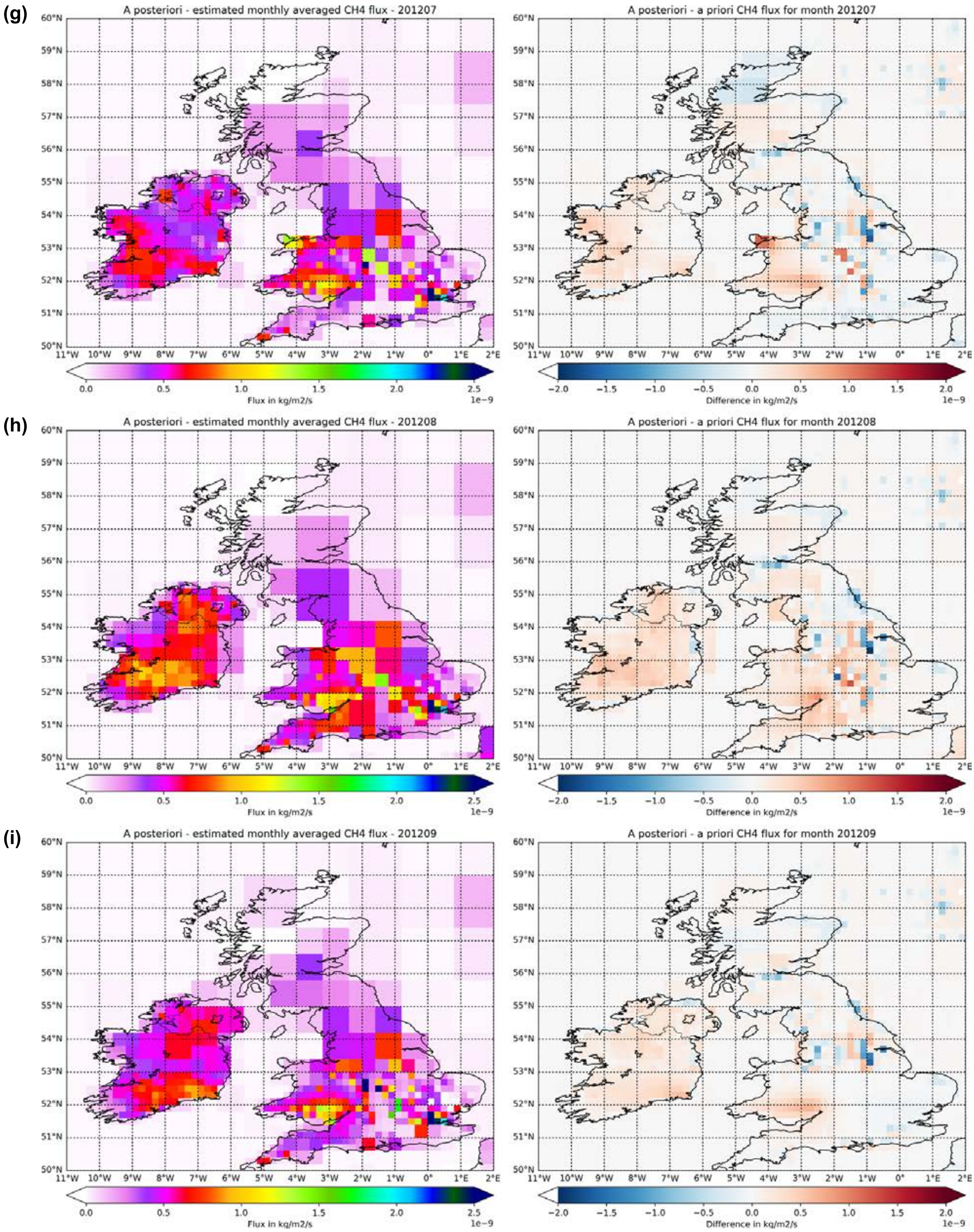

Figure 4.23. Continued. (g) July; (h) August; (i) September.

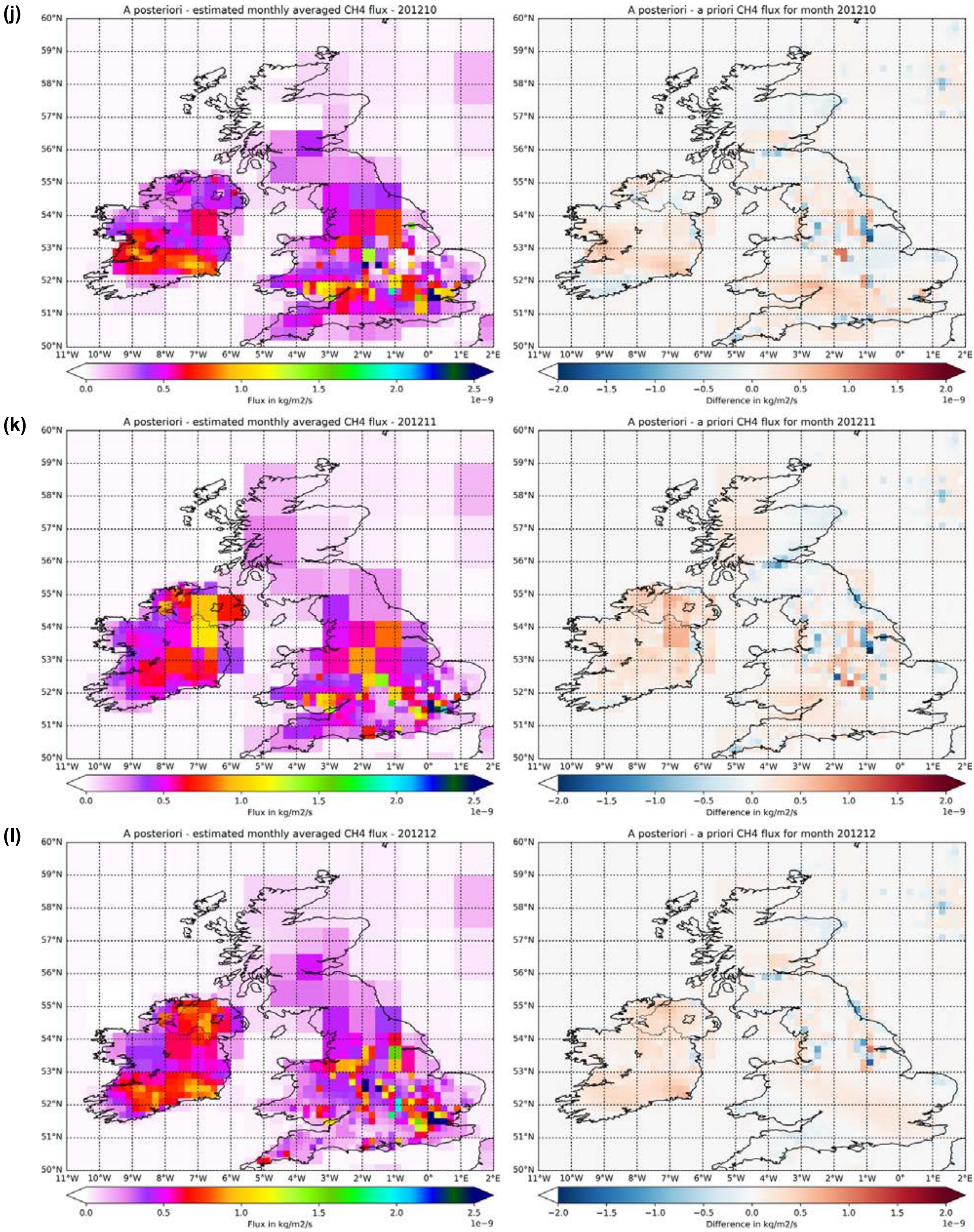

Figure 4.23. Continued. (j) October; (k) November; and (l) December.

northern areas of Ulster. Negative corrections occur in northern Connacht and eastern Ulster. In June (see Figure 4.23f), the corrections are again mild: positive in the centre of Ireland and negative to the south and in Dublin. July (see Figure 4.23g) shows no correction in most areas of Ulster and Leinster and a general positive correction in the west. August (see Figure 4.23h) shows an overall large positive correction in Ireland, except for the Dublin region and the southern tip of Munster, with large positive corrections in the upper regions of Munster province. September (see Figure 4.23i) shows relatively mild positive corrections in the fluxes with respect to the a priori data in Ireland. Positive corrections of a similar magnitude occur in October (see Figure 4.23j) in central Ireland, with a large area of negative corrections in the south of Munster. November (see Figure 4.23k) shows an area with aggregated output in northern Leinster, with a large increase in the flux. December (see Figure 4.23l) again shows a general upwards correction of the a priori fluxes.

There is no clear pattern in the overall variation in total emissions other than a tendency for higher emissions in the summer months (Figure 4.24). This analysis is interesting and merits an extended analysis of multiple years of data, to establish if seasonal trends can be identified.

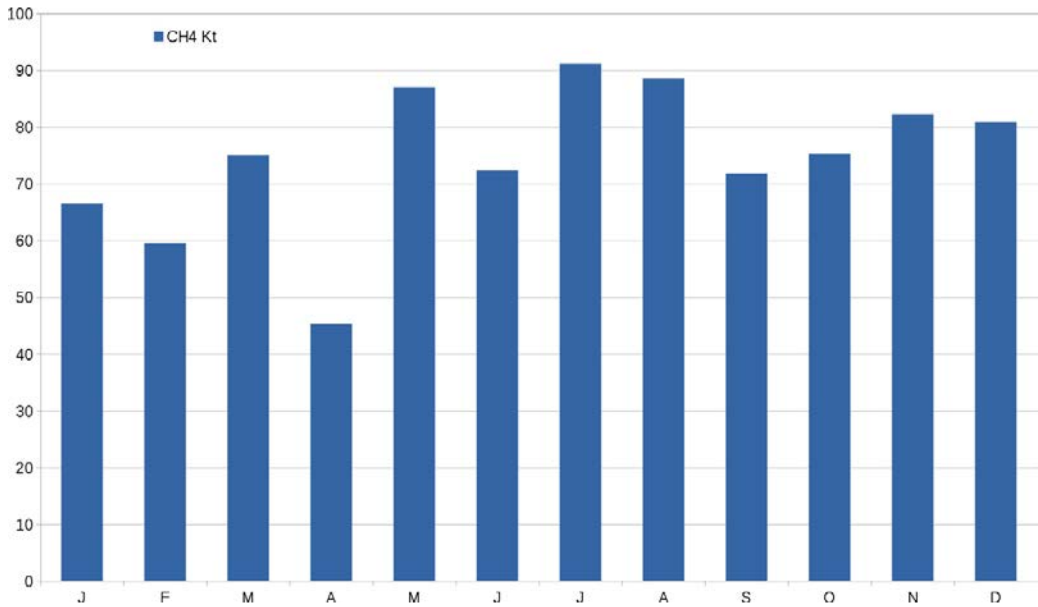

Figure 4.24. Monthly total estimated Irish emissions in kt.

5 Conclusions

The IMPliCIt project has taken the first steps to increase the capabilities of Ireland in the field of inverse modelling of GHGs, in particular CH_4. The IMPLiCIT project has implemented and tested the FLEXINVERT Bayesian inversion framework using different sets of observational data and a priori information combined with atmospheric transport modelling calculations made with the widely used FLEXPART model. FLEXINVERT has been adapted and prepared for the Irish domain, with minor adaptations of the code and the preparation of the programming environment to arrange and perform the necessary runs and the corresponding outcome evaluation. Initial inversion estimates indicate slightly higher inferred emissions than the a priori estimate (~30%). These estimates are likely to be on the margin of the uncertainties associated with both bottom-up and top-down methods. It is concluded that additional work is needed to contextualise the results using a multiyear study and to evaluate the different contributors to the uncertainty and identify relevant procedures to quantify and incorporate these uncertainties into estimates.

6 Recommendations

It is clear from the work performed in the IMPLiCIt project that additional actions are required to understand and bridge the gap between the fluxes estimated through a bottom-up approach, and used in the nationally provided total emissions, and those estimated through a top-down approach. The following recommendations are made (Figure 6.1):

- Perform multiyear, high temporal resolution analysis to explore the inter-annual and seasonal variation in flux estimates. This would include spike removal on all datasets to preclude any local interference.
- Further investigate the sensitivity of the inversion system to different meteorological driving data, background definition, a priori data and boundary layer height in the model.
- Constrain the uncertainties related to the atmospheric transport modelling (calculation of the SRSs) through ensemble modelling.
- Explore additional measurement sites that would allow, from a sectoral perspective, a better comparison with bottom-up emissions inventories.
- Continue to enhance this newly developed capacity to deliver improved flux estimations, which is essential for the delivery of more accurate and verified national emissions inventories for Ireland.
- Upgrade the observational network to include routine isotopic composition measurements to further constrain emission sectors.

In addition, closer co-operation and collaboration with the producers of bottom-up inventories, to jointly address discrepancies, would be most beneficial.

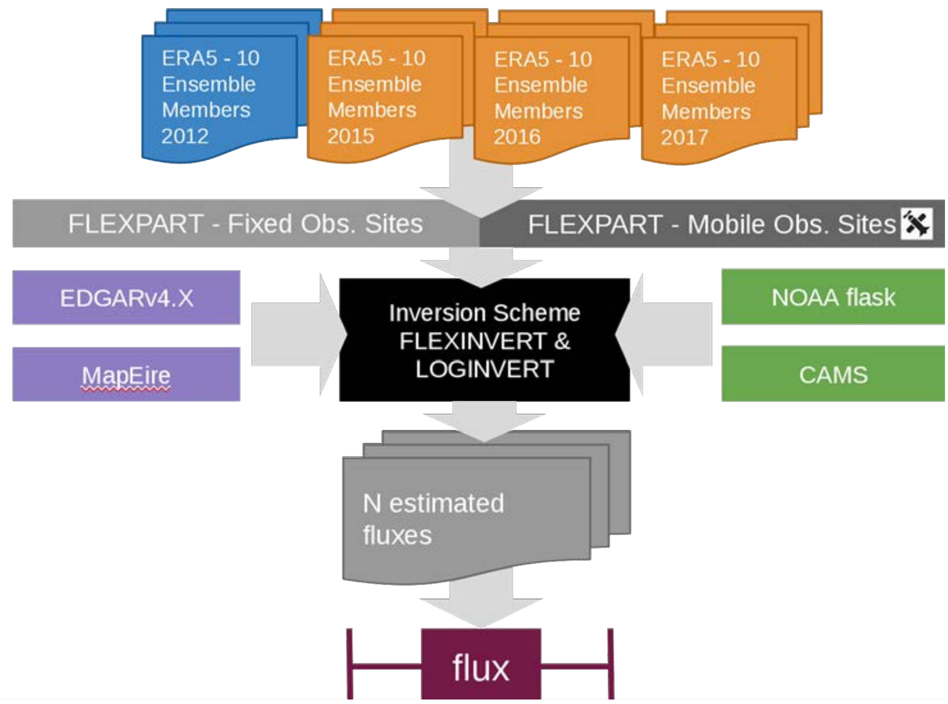

Figure 6.1. Additional activities to be performed in order to obtain better estimates of the emissions inventories using a top-down approach and to bridge the gap with the bottom-up assessments. CAMS, Copernicus Atmospheric Monitoring Service; ERA, ECMWF Re-analysis.

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Abbreviations

CTM	Chemical transport model
ECMWF	European Centre for Medium-Range Weather Forecasts
EDGAR	Emissions Database for Global Atmospheric Research
EPA	Environmental Protection Agency
FLEXINVERT	FLEXible INVERTion dispersion model
FLEXPART	FLEXible PARTicle dispersion model
GHG	Greenhouse gas
GNFR	Goods not for resale
IMPLiCIt	IMProving inversion modeL Capability in Ireland
InGOS	Integrated non-CO_2 Greenhouse gas Observing System
LPDM	Lagrangian particle dispersion model
LULUCF	Land use, land-use change and forestry
NFR	Norwegian Research Council
NILU	Norwegian Institute for Air Research
NOAA	National Oceanic and Atmospheric Administration
NSD	Normalised standard deviation
RMSE	Root mean square error
SRS	Source receptor sensitivity
TM5/TM65	Topographic Mapping
WDCGG	World Data Centre for Greenhouse Gases
WRF	Weather Research and Forecasting

AN GHNÍOMHAIREACHT UM CHAOMHNÚ COMHSHAOIL
Tá an Ghníomhaireacht um Chaomhnú Comhshaoil (GCC) freagrach as an gcomhshaol a chaomhnú agus a fheabhsú mar shócmhainn luachmhar do mhuintir na hÉireann. Táimid tiomanta do dhaoine agus don chomhshaol a chosaint ó éifeachtaí díobhálacha na radaíochta agus an truaillithe.

Is féidir obair na Gníomhaireachta a roinnt ina trí phríomhréimse:

Rialú: *Déanaimid córais éifeachtacha rialaithe agus comhlíonta comhshaoil a chur i bhfeidhm chun torthaí maithe comhshaoil a sholáthar agus chun díriú orthu siúd nach gcloíonn leis na córais sin.*

Eolas: *Soláthraímid sonraí, faisnéis agus measúnú comhshaoil atá ar ardchaighdeán, spriocdhírithe agus tráthúil chun bonn eolais a chur faoin gcinnteoireacht ar gach leibhéal.*

Tacaíocht: *Bímid ag saothrú i gcomhar le grúpaí eile chun tacú le comhshaol atá glan, táirgiúil agus cosanta go maith, agus le hiompar a chuirfidh le comhshaol inbhuanaithe.*

Ár bhFreagrachtaí

Ceadúnú
Déanaimid na gníomhaíochtaí seo a leanas a rialú ionas nach ndéanann siad dochar do shláinte an phobail ná don chomhshaol:
- saoráidí dramhaíola *(m.sh. láithreáin líonta talún, loisceoirí, stáisiúin aistrithe dramhaíola)*;
- gníomhaíochtaí tionsclaíocha ar scála mór *(m.sh. déantúsaíocht cógaisíochta, déantúsaíocht stroighne, stáisiúin chumhachta)*;
- an diantalmhaíocht *(m.sh. muca, éanlaith)*;
- úsáid shrianta agus scaoileadh rialaithe Orgánach Géinmhodhnaithe *(OGM)*;
- foinsí radaíochta ianúcháin *(m.sh. trealamh x-gha agus radaiteiripe, foinsí tionsclaíocha)*;
- áiseanna móra stórála peitril;
- scardadh dramhuisce;
- gníomhaíochtaí dumpála ar farraige.

Forfheidhmiú Náisiúnta i leith Cúrsaí Comhshaoil
- Clár náisiúnta iniúchtaí agus cigireachtaí a dhéanamh gach bliain ar shaoráidí a bhfuil ceadúnas ón nGníomhaireacht acu.
- Maoirseacht a dhéanamh ar fhreagrachtaí cosanta comhshaoil na n-údarás áitiúil.
- Caighdeán an uisce óil, arna sholáthar ag soláthraithe uisce phoiblí, a mhaoirsiú.
- Obair le húdaráis áitiúla agus le gníomhaireachtaí eile chun dul i ngleic le coireanna comhshaoil trí chomhordú a dhéanamh ar líonra forfheidhmiúcháin náisiúnta, trí dhíriú ar chiontóirí, agus trí mhaoirsiú a dhéanamh ar leasúchán.
- Cur i bhfeidhm rialachán ar nós na Rialachán um Dhramhthrealamh Leictreach agus Leictreonach (DTLL), um Shrian ar Shubstaintí Guaiseacha agus na Rialachán um rialú ar shubstaintí a ídíonn an ciseal ózóin.
- An dlí a chur orthu siúd a bhriseann dlí an chomhshaoil agus a dhéanann dochar don chomhshaol.

Bainistíocht Uisce
- Monatóireacht agus tuairisciú a dhéanamh ar cháilíocht aibhneacha, lochanna, uiscí idirchriosacha agus cósta na hÉireann, agus screamhuiscí; leibhéil uisce agus sruthanna aibhneacha a thomhas.
- Comhordú náisiúnta agus maoirsiú a dhéanamh ar an gCreat-Treoir Uisce.
- Monatóireacht agus tuairisciú a dhéanamh ar Cháilíocht an Uisce Snámha.

Monatóireacht, Anailís agus Tuairisciú ar an gComhshaol
- Monatóireacht a dhéanamh ar cháilíocht an aeir agus Treoir an AE maidir le hAer Glan don Eoraip (CAFÉ) a chur chun feidhme.
- Tuairisciú neamhspleách le cabhrú le cinnteoireacht an rialtais náisiúnta agus na n-údarás áitiúil *(m.sh. tuairisciú tréimhsiúil ar staid Chomhshaol na hÉireann agus Tuarascálacha ar Tháscairí)*.

Rialú Astaíochtaí na nGás Ceaptha Teasa in Éirinn
- Fardail agus réamh-mheastacháin na hÉireann maidir le gáis cheaptha teasa a ullmhú.
- An Treoir maidir le Trádáil Astaíochtaí a chur chun feidhme i gcomhair breis agus 100 de na táirgeoirí dé-ocsaíde carbóin is mó in Éirinn.

Taighde agus Forbairt Comhshaoil
- Taighde comhshaoil a chistiú chun brúnna a shainaithint, bonn eolais a chur faoi bheartais, agus réitigh a sholáthar i réimsí na haeráide, an uisce agus na hinbhuanaitheachta.

Measúnacht Straitéiseach Timpeallachta
- Measúnacht a dhéanamh ar thionchar pleananna agus clár beartaithe ar an gcomhshaol in Éirinn *(m.sh. mórphleananna forbartha)*.

Cosaint Raideolaíoch
- Monatóireacht a dhéanamh ar leibhéil radaíochta, measúnacht a dhéanamh ar nochtadh mhuintir na hÉireann don radaíocht ianúcháin.
- Cabhrú le pleananna náisiúnta a fhorbairt le haghaidh éigeandálaí ag eascairt as taismí núicléacha.
- Monatóireacht a dhéanamh ar fhorbairtí thar lear a bhaineann le saoráidí núicléacha agus leis an tsábháilteacht raideolaíochta.
- Sainseirbhísí cosanta ar an radaíocht a sholáthar, nó maoirsiú a dhéanamh ar sholáthar na seirbhísí sin.

Treoir, Faisnéis Inrochtana agus Oideachas
- Comhairle agus treoir a chur ar fáil d'earnáil na tionsclaíochta agus don phobal maidir le hábhair a bhaineann le caomhnú an chomhshaoil agus leis an gcosaint raideolaíoch.
- Faisnéis thráthúil ar an gcomhshaol ar a bhfuil fáil éasca a chur ar fáil chun rannpháirtíocht an phobail a spreagadh sa chinnteoireacht i ndáil leis an gcomhshaol *(m.sh. Timpeall an Tí, léarscáileanna radóin)*.
- Comhairle a chur ar fáil don Rialtas maidir le hábhair a bhaineann leis an tsábháilteacht raideolaíoch agus le cúrsaí práinnfhreagartha.
- Plean Náisiúnta Bainistíochta Dramhaíola Guaisí a fhorbairt chun dramhaíl ghuaiseach a chosc agus a bhainistiú.

Múscailt Feasachta agus Athrú Iompraíochta
- Feasacht chomhshaoil níos fearr a ghiniúint agus dul i bhfeidhm ar athrú iompraíochta dearfach trí thacú le gnóthais, le pobail agus le teaghlaigh a bheith níos éifeachtúla ar acmhainní.
- Tástáil le haghaidh radóin a chur chun cinn i dtithe agus in ionaid oibre, agus gníomhartha leasúcháin a spreagadh nuair is gá.

Bainistíocht agus struchtúr na Gníomhaireachta um Chaomhnú Comhshaoil
Tá an ghníomhaíocht á bainistiú ag Bord lánaimseartha, ar a bhfuil Ard-Stiúrthóir agus cúigear Stiúrthóirí. Déantar an obair ar fud cúig cinn d'Oifigí:
- An Oifig um Inmharthanacht Comhshaoil
- An Oifig Forfheidhmithe i leith cúrsaí Comhshaoil
- An Oifig um Fianaise is Measúnú
- Oifig um Chosaint Radaíochta agus Monatóireachta Comhshaoil
- An Oifig Cumarsáide agus Seirbhísí Corparáideacha

Tá Coiste Comhairleach ag an nGníomhaireacht le cabhrú léi. Tá dáréag comhaltaí air agus tagann siad le chéile go rialta le plé a dhéanamh ar ábhair imní agus le comhairle a chur ar an mBord.

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IMPLiCIt: IMProving inversion modeL Capability in Ireland

Authors: Colin O'Dowd, Damien Martin and Dèlia Arnold

The IMPLICIT project builds national capacity in the area of climate change and, in particular, extends our understanding of national greenhouse gas emissions using in situ observations and state-of-the-art computer modelling to produce top-down national estimates and verification of bottom-up traditional estimates.

Identifying Pressures

Irish greenhouse gas emissions are internationally reported and are based on statistical activity data comprising source-specific and country-specific emission factors. For methane and nitrous oxide, however, such "bottom-up" emission inventories have a substantial degree of uncertainty, mainly because of variability of emission factors and the influence of natural sources and processes. This project aims to constrain these uncertainties with a complementary top-down approach using inverse modelling techniques and observational data from Ireland's climate change monitoring network.

Informing Policy

Ireland's 2020 target is to achieve a 20% reduction in non-Emission Trading Scheme (non-ETS) sector emissions (i.e. agriculture, transport, the built environment, waste and non-energy-intensive industry) relative to 2005 levels, with annual limits set for each year over the period 2013–2020. This project informs the efficacy of proposed mitigation strategies by producing annual mapped national emission estimates for greenhouse gases, with a special focus on methane.

Developing Solutions

A solution to the pressing issue of greenhouse gases is to develop national capability in terms of inversion model capability and this project seeks to deliver the first steps in this development. The FLEXINVERT modelling system can be deployed to produce inversions for any species for which atmospheric loss (if any) can be described as a linear process, such as radioactive decay, dry and wet deposition, and oxidative chemistry. Furthermore, the modelling framework described can be used on a range of scales, including continental, regional and local, and, as well as methane, can be further utilised for nitrous oxide, carbon dioxide and other atmospheric species, offering the possibility of constraining a range of emissions estimates, given suitable measurements.

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